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LAWRENCE LIVERMORE NATIONAL LABORATORY

LDRD Highlights at the National Laboratories

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October 14, 2016

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U.S. DEPARTMENT OF ENERGY

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LDRD

Laboratory Directed Research and Development at the National Laboratories

Highlights

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Laboratory Directed Research and Development

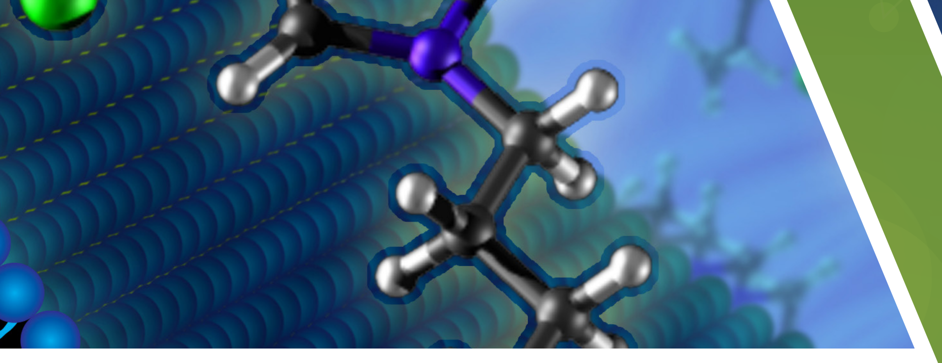
Enabling the national laboratories to address tomorrow's biggest challenges

To meet the nation's critical challenges, the Department of Energy (DOE) national laboratories have always pushed the boundaries of science, technology, and engineering. The Atomic Energy Act of 1954 provided the basis for these laboratories to engage in the cutting edge of science and technology and respond to technological surprises, while retaining the best scientific and technological minds. To help re-energize this commitment, in 1991 the U.S. Congress authorized the national laboratories to devote a relatively small percentage of their budget to creative and innovative work that serves to maintain their vitality in disciplines relevant to DOE missions. Since then, this effort has been formally called the Laboratory Directed Research and Development (LDRD) Program. LDRD has been an essential mechanism to enable the laboratories to address DOE's current and future missions with leading-edge research proposed independently by laboratory technical staff, evaluated through expert peer-review committees, and funded by the individual laboratories consistent with the authorizing legislation and the DOE LDRD Order 413.2C.

Today, that work continues to be critical. LDRD researchers are exploring new ideas in fundamental scientific issues that include energy, environmental, and nuclear security. As DOE Secretary of Energy Ernest Moniz has affirmed in his annual report to Congress, "the LDRD Program provides the laboratories with the opportunity and flexibility to establish and maintain an environment that encourages and supports creativity and innovation, and contributes to their long-term viability. LDRD allows the Department's laboratories to position themselves to advance our national security mission and respond to our Nation's future research needs."

Accelerating the pace of scientific and technological innovation requires engaging the brightest researchers and creating the necessary advanced computational and experimental tools. LDRD is helping the nation lead basic research in the physical sciences and advance foundational sciences through development of the next generation of experimental, characterization, and computational tools, as well as distinctive scientific instrumentation. These capabilities and tools, when fully developed, can also be made available to academia, industry, and other scientists nationally to help advance DOE's mission and accelerate technological innovations. Furthermore, LDRD investment in undergraduate and graduate students, postdoctoral researchers, and scientific collaborations leads the way to developing new science, technology, and engineering talent, while simultaneously paving the path for solutions and innovations through potentially high-payoff research and development projects.

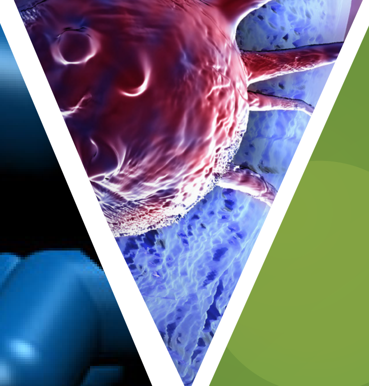
This publication highlights some of the cutting-edge research that is pushing the envelope in scientific and engineering fields critical to addressing the DOE strategy. These include projects that address our energy and environmental needs and nuclear security concerns, as well as promoting U.S. leadership in scientific discovery and innovation. While each project is impressive in its own right, it is even more important to remember each project is part of a much-larger portfolio that helps keep the national laboratories ready to deal with tomorrow's critical challenges.

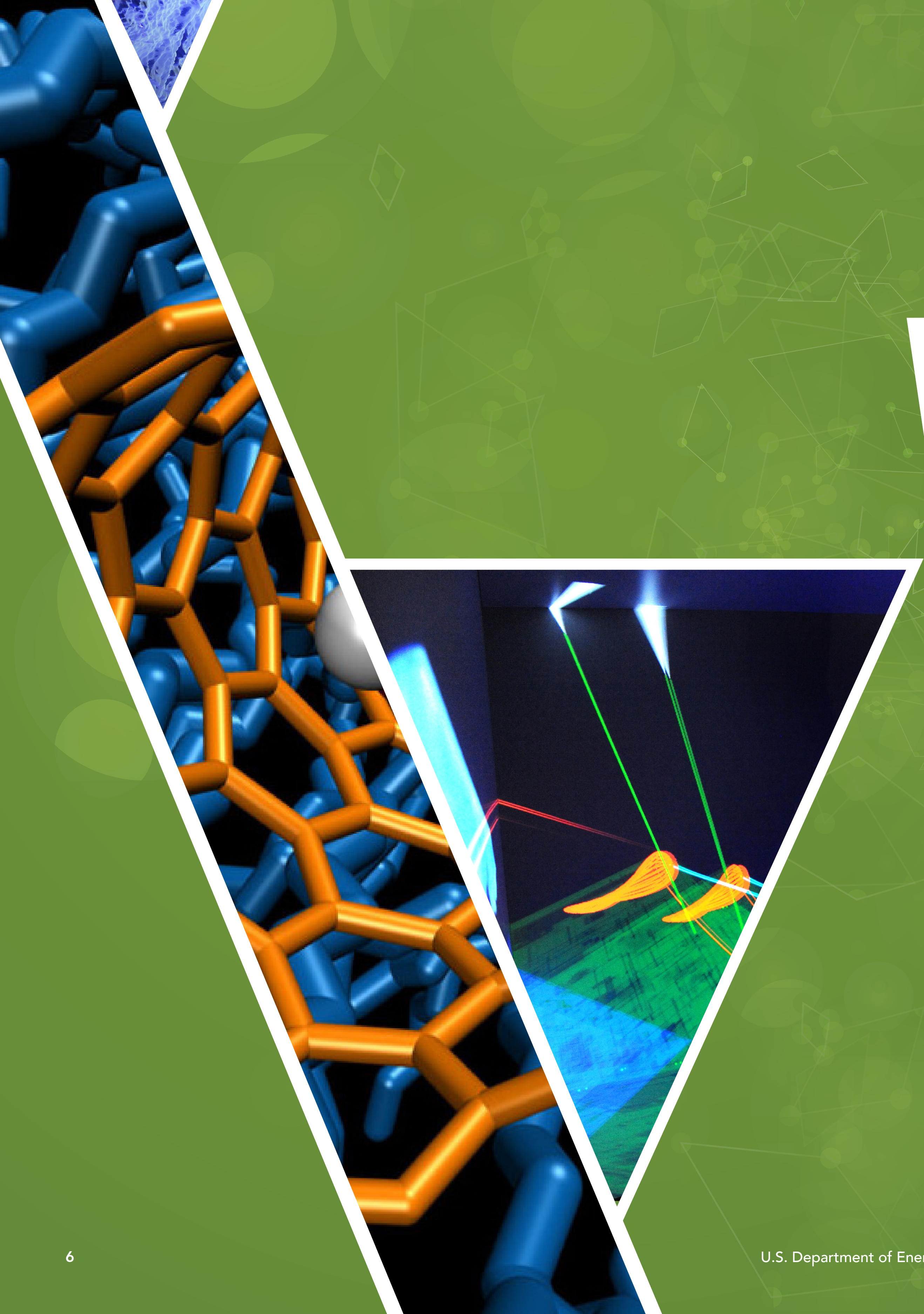


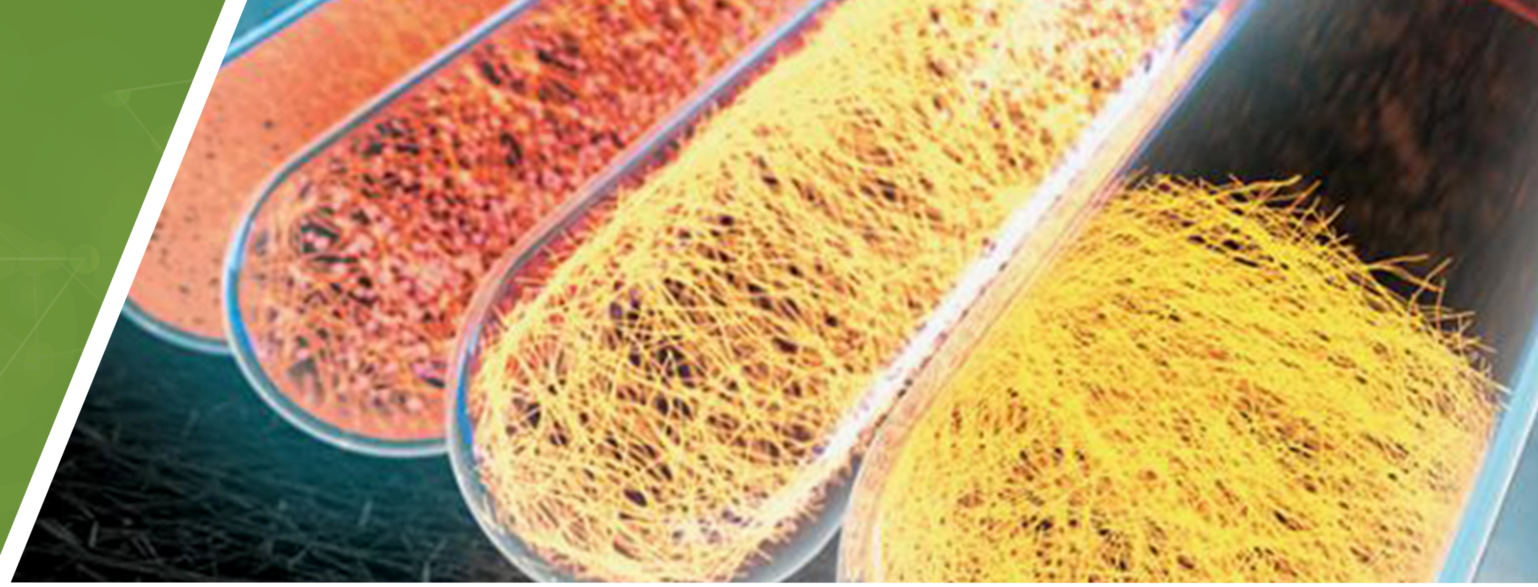
ENERGY & ENVIRONMENTAL SECURITY

Innovative and transformative LDRD efforts are leading the way to reduce America's dependence on oil with an "all of the above" strategy to improve energy security, increase the nation's economic competitiveness, and cut carbon pollution. In addition, LDRD is working to ensure environmental security through remediation efforts and by detecting and responding to natural or human-caused biothreats.

LDRD projects are helping to achieve the nation's aggressive goal of generating 80% of our electricity from clean energy sources (including nuclear) by the year 2035, and reducing carbon pollution by 3 billion metric tons. LDRD research for these efforts includes improved clean-energy technologies, smart-grid solutions for integrated multiple energy sources, advanced energy-storage technologies, advanced transportation and manufacturing capabilities, enhanced waste-reduction technologies, and efficient water usage and recycling technologies. Implicit in these challenges, to accelerate discovery and innovation, is the requirement to integrate modeling, simulation, and experimental capabilities for designing and developing novel, high-performance materials with applications in solar resources, energy storage, clean water production, environmental remediation, and molecular-to-systems-scale studies to address energy and climate challenges. LDRD projects, for example, are helping to develop solutions leading to improved high-performance solar cells, new energy storage technologies, advanced energy-efficient manufacturing processes, a nuclear fuel cycle with increased efficiency (reduced waste), and an integrated, efficient, and resilient electric grid. Additionally, LDRD researchers are seeking ways to lower human and environmental risk by developing highly effective groundwater remediation of key radionuclides, as well as detecting the nature of biothreats with a smartphone microscope.







ENERGY & ENVIRONMENTAL SECURITY

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Materials Discovery Opens Pathway Toward a New Kind of Battery

Argonne National Laboratory

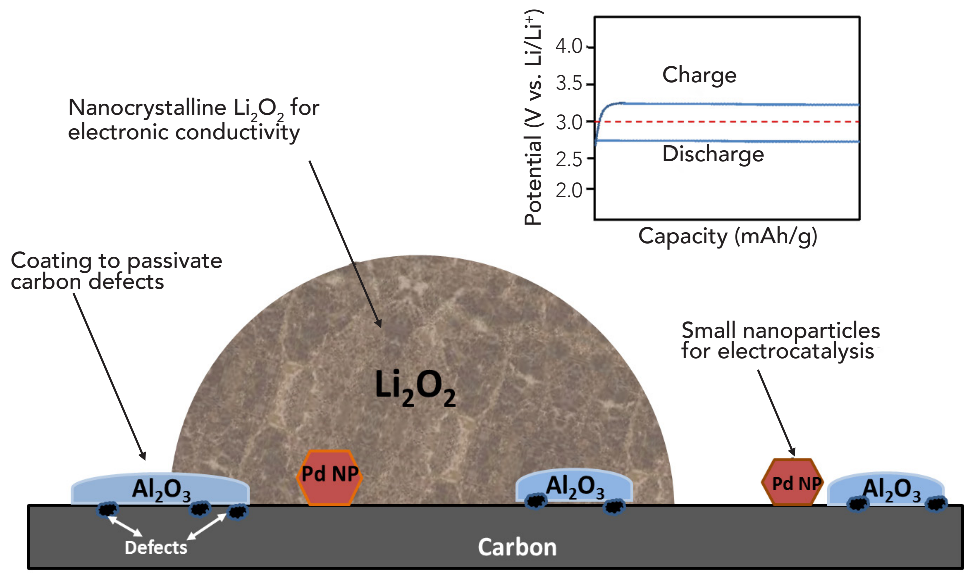
Efficiency of lithium–oxygen batteries enhanced by metallic nanoparticles

Project Overview

Lithium–oxygen ($Li–O_2$) batteries have the potential for higher energy density than conventional lithium-ion batteries. However, when charging the battery, the large voltage (or overpotential) required to decompose the products that had formed at the battery's cathode during battery discharge limits performance. Researchers at Argonne National Laboratory have discovered new cathode material architectures that can significantly reduce the voltage required to decompose those products. The new architectures' use of metal nanoparticles enables better cathode performance.

Why It Matters

New energy storage technologies in general, and development of new kinds of batteries in particular, are a principal goal of DOE that is pursued vigorously at Argonne. One major use envisioned for a future efficient $Li–O_2$ battery is powering long-range electric vehicles. Overcoming current challenges such as poor cycle life and large charge overpotentials is the goal of this research, which draws upon Argonne's capabilities in theory, advanced materials, and a long record of battery development. The way in which the $Li–O_2$ battery works is fundamentally different from lithium-ion batteries because it uses chemical transformations that result in the growth of a new bulk phase on its cathode rather than intercalation of lithium into a metal oxide cathode. This is what enables $Li–O_2$ batteries to have much greater potential for high-energy density. Khalil Amine, head of the Technology Development Group in Argonne's Battery Technology Department, explains, "solving the challenges remaining for the development of $Li–O_2$ batteries relies on a multidisciplinary approach well suited to a national laboratory such as Argonne, therefore advancing fundamental studies of materials at the same time the technology is advanced." For the $Li–O_2$ problem, that approach resulted in new materials architectures based on nanoparticles that reduce the energy required to break down the bulk lithium peroxide (Li_2O_2) phase formed on the cathode.



Schematic of an Al_2O_3 coated carbon surface, with palladium nanoparticles and nanocrystalline lithium peroxide, which contribute to a dramatically more efficient $Li–O_2$ battery. The inset shows a hypothetical charge/discharge profile.

Technical & Scientific Achievements

Amine noted that "two new cathode architectures based on nanoscale components were discovered." They resulted in a dramatic reduction in charge overpotential and an increase in efficiency of the $Li–O_2$ battery. In one case the cathode utilizes atomic layer deposition of palladium nanoparticles on a carbon surface with an alumina coating for passivation of carbon defect sites. The low charge potential is enabled by the combination of palladium nanoparticles attached to the carbon cathode surface, a nanocrystalline form of Li_2O_2 with grain boundaries, and the alumina coating preventing electrolyte decomposition on carbon. High-resolution transmission electron microscopy provided evidence for the nanocrystalline form of Li_2O_2. Better pathways for conducting electrons to where the surface meets the electrolyte allow for easier decomposition on charging of the battery.

The second cathode material that was developed was based on stabilization of crystalline LiO_2 in a $Li–O_2$ battery for the first time. This was accomplished using iridium nanoparticles on a graphene-based cathode. Only LiO_2 is seen on the cathode with no evidence of Li_2O_2. A novel templating growth mechanism involving the iridium nanoparticles on the cathode surface may be responsible for the growth of crystalline LiO_2. Project results demonstrate that LiO_2 formed in the $Li–O_2$ battery is stable enough for the battery to be repeatedly charged and discharged with a very low charge potential.

Contributions & Impact

The new cathode architectures based on palladium and iridium nanoparticles open promising new avenues for $Li–O_2$ battery development through optimization of the metal nanoparticle and the carbon support, as well as incorporation of new protective coatings. These techniques are currently enabling improvement of efficiency and cycle life of $Li–O_2$ batteries. The new cathode material architectures will usher in a new regime of advanced batteries that have significantly greater energy density than today's lithium-ion batteries and that will ultimately lead to long-range electric vehicles. The potential for application of the $Li–O_2$ battery to other electrical energy storage needs also bears further investigation. This work was one of the precursors to the advent of DOE's Energy Innovation Hub, the Joint Center for Energy Storage Research, led by Argonne, which brings to bear the resources and expertise of several laboratories and industry to address challenges such as those presented by the $Li–O_2$ case.

This work resulted in 16 invention report disclosures, 12 patent applications, and 4 patents.

Publications

Lu, J., et al., "A Lithium–Oxygen Battery Based on Lithium Superoxide." *Nature* **529**(7586), 377–382 (2016).

Lu, J., et al., "Effect of the Size-Selective Silver Clusters on Lithium Peroxide Morphology in Lithium–Oxygen Batteries." *Nat. Comm.* **5**, 4895 (2014).

Lu, J., et al., "A Nanostructured Cathode Architecture for Low Charge Overpotential in Lithium–Oxygen Batteries." *Nat. Comm.* **4,** 2383 (2013).

Trahey, L., et al., "Synthesis, Characterization, and Structural Modeling of High-Capacity, Dual Functioning MnO_2 Electrode/Electrocatalysts for $Li-O_2$ Cells." *Adv. Eng. Mater.* **3**(1), 75–84 (2013).

Zhai, D., et al., "Disproportionation in $Li–O_2$ Batteries Based on a Large Surface Area Carbon Cathode." *J. Am. Chem. Soc.* **135**(41), 15364–15372 (2013).

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Acknowledgments

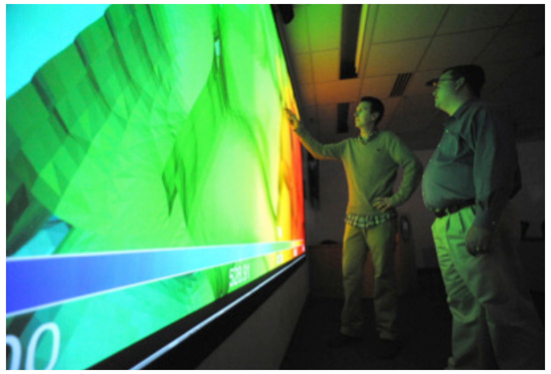
This work was the result of research carried out by a large team of multidisciplinary scientists at Argonne led by K. Amine, M. Thackeray, and L. A. Curtiss. External collaborators included J. Pan (University of South Florida), G. W. Feigenson (Cornell University), and G. Pabst (University of Graz).

"The new architectures' use of metal nanoparticles enables better cathode performance."

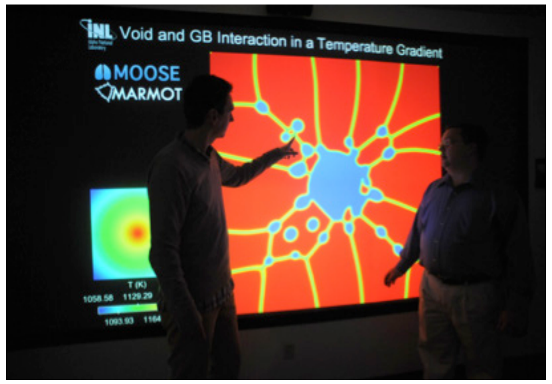
MOOSE Capability Extension in Support of Full-Core Modeling

Idaho National Laboratory

Adding fundamental capabilities to the computer simulation framework known as MOOSE in support of multiscale reactor modeling



Derek Gaston (left) and Michael Tonks (right) look at a simulation result on the powerwall.



Derek Gaston and Michael Tonks view fission gas bubbles from a MARMOT mesoscale fuel performance code simulation.

Project Overview

Nuclear reactor operation involves multiscale, multiphysics phenomena where large-scale physics, such as fluid flow and heat conduction, are driven by atomistic effects of the fission process. This LDRD project aimed to simplify the modeling and simulation of such complex system. Researchers at Idaho National Laboratory created a pluggable system for running multiple applications in a "tightly coupled" or "loosely coupled" strategy, as opposed to the default "fully coupled" strategy of MOOSE (Multiphysics Object Oriented Simulation Environment). MultiApps, Transfers, and Restart systems were added to enable independent physics simulations to run in concert without significantly increasing the memory footprint of the underlying numerical solvers. The enhancements enabled a first-of-a-kind, full-core, multiscale nuclear reactor simulation through an efficient linking method.

Why It Matters

In the effort to extend the life of the nation's reactor fleet while continuing to make operations safer, industry and the DOE face many challenges that require significant advances in the accuracy and fidelity of modeling and simulation technologies. The capabilities added through this LDRD enable whole new classes of simulations not currently feasible with competing frameworks. Examples include the coupling of multiscale systems to accurately capture highly nonlinear effects between scales, or the modeling of reactor accident scenarios in which the state of the fuel and cooling systems impacts the progression of the accident.

Technical & Scientific Achievements

This project was highlighted by large advances in software engineering practices to accelerate development of full-core simulation capabilities. "In March 2014, MOOSE was released as open-source software, which has led to a rapid increase in both the number of its users and the number of its developers," relates co-investigator Cody Permann. MOOSEBuild, a new tool for coordinating development of multiple, interdependent MOOSE-based applications, was also created. During its second year, the MOOSE project was awarded an R&D 100 Award from *R&D Magazine* in large part because of the full-core simulation capability developed during this LDRD.

The Consortium for Advanced Simulation of Light Water Reactors project is now making extensive use of the MultiApps system for its virtual reactor project. New capabilities to dynamically link applications together "on the fly" have also been implemented, further enabling advanced multi-application simulations. Finally, the lessons learned from the original development of the tool MOOSEBuild (the continuous integration testing utility) have been used to develop an improved, second-generation tool. This tool, which is currently called Civet, can interact with more open-source tools, is more flexible for the main projects based on the Git version-control system used for software hosted by Idaho National Laboratory, and contains an improved user interface. Civet will eventually be made open-source and used for external scientific development workflows.

Contributions & Impact

This project has benefited several DOE missions. It directly benefits energy resources because the capabilities produced are already being utilized to study advanced nuclear reactor and fuel concepts. The MOOSE platform is currently being leveraged extensively by both the Nuclear Engineering Advanced Modeling and Simulation and the Consortium for Advanced Simulation of Light Water Reactors programs. These capabilities are also impacting environmental quality and science. The intrinsic capabilities being developed are being applied to many disparate areas of environmental science and scientific endeavors (including fundamental material science studies).

Publications

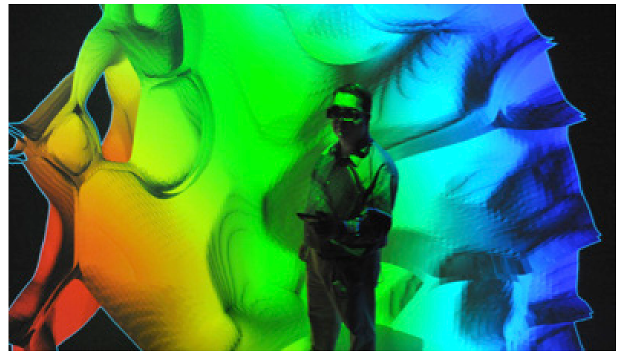
Gaston, D. R., et al., "Physics-Based Multiscale Coupling for Full Core Nuclear Reactor Simulation," *Ann. Nucl. Energ.*, *Special Issue on Multi-Physics Modelling of LWR Static and Transient Behaviour* **84**, 45–54 (2015).

Mangeri, J., et al., "Influence of Elastic and Surface Strains on the Optical Properties of Semi-Conducting Core-Shell Nanoparticles," *Phys. Rev. Appl.* **4**(1), 014001–014011 (2015).

Short, M., et al., "A Perspective on Coupled Multiscale Simulation and Validation in Nuclear Materials." *MRS Bull.* **39**(1), 71–77 (2014).

Acknowledgments

This work was led by principal investigator Derek Gaston of Idaho National Laboratory. Co-investigators included Cody Permann, David Andrs, and John Peterson from Idaho National Laboratory, and Dmitry Karpeev from Argonne National Laboratory.



Michael Tonks explores a MARMOT mesoscale fuel performance code result in the CAVE 3D computer-assisted virtual environment.

Jason Miller controls the powerwall from the crow's nest.

"Large-scale physics, such as fluid flow and heat conduction, are driven by atomistic effects of the fission process."

New Processing Methods Enable High-Performance, Perovskite-Based Solar Cells

National Renewable Energy Laboratory

Novel solution-deposition approaches for perovskite solar cells

Project Overview

This project at the National Renewable Energy Laboratory developed new synthetic approaches for making high-quality, uniform films of the new perovskite material that exploded on the solar scene in 2009. The new materials and processing approaches developed in this project enable solution-processed photovoltaics that promise to combine good materials utilization and high efficiency with lower cost, and therefore will help make possible the terawatt deployment of photovoltaics.

Why It Matters

Solution-processed solar cells have the ability to reduce the cost of solar panels, thus making renewable electricity outpower traditional polluting technologies. The work in this project developed new tools and expertise in perovskite photovoltaics that will drive the technology toward rapid commercialization. For example, understanding the processing chemistry and control of film morphology is essential towards learning how to scale up this technology, eventually reaching terawatt scales at costs so low DOE could actually exceed its SunShot initiative goal to reduce the total installed cost of solar systems to $0.06 per kilowatt-hour by 2020. With the rapid rise in efficiency, the field of perovskite photovoltaics now must tackle the challenge of making the perovskite solar cells stable enough to last for many years, similar to existing silicon technology.

Technical & Scientific Achievements

Much of the underlying chemistry and physics of the specific absorber (methylammonium lead triiodide) was practically unstudied before 2009. The perovskite-structure material contains an organic molecule housed within a lead halide cage. Initial devices using this material were fabricated using a liquid electrolyte as one contact, which severely limited device lifetime. The efforts were later shifted toward devices containing solid-state contacting layers. This project had made significant advances in developing the synthetic approaches for making high-quality, uniform halide perovskite films with controlled morphology (e.g., crystal phase, grain size, and crystal orientation) and composition (e.g., mixed anions and cations). The control of the morphology and composition of the films is enabled by controlling the precise chemistry of the precursor solutions and resulted in much more reproducible and robust perovskite films. These new approaches were used to develop high-performance, solid-state perovskite solar cells. Leveraging National Renewable Energy Laboratory's existing core capabilities in photovoltaics research and development as well as in fundamental research, this project obtained valuable information on the impact of different film morphologies and device architectures on charge carrier dynamics and device characteristics.

Contributions & Impact

This LDRD project propelled the field in a major way by producing 9 peer-reviewed journal articles, including 1 invited perspective, 10 records of invention, and 4 patent/provisional patent applications. The publications and patents concerned new synthetic approaches for making high-quality, uniform halide perovskite films with controlled morphology and composition that are enabling large-scale, roll-to-roll deposited solar cells. In one part of the project, researchers demonstrated a new two-step process that speeds up the formation of the perovskite film by a factor of 10, significantly increasing the reproducibility and reliability of the films. Next to precipitating new projects within DOE's Solar Energy Technologies office, it has also helped initiate work with industrial partners that is rapidly pushing perovskite solar cell technology towards commercialization.

Publications

Zhang, T., et al., "Controllable Sequential Deposition of Planar $CH_3NH_3PbI_3$ Perovskite Films via Adjustable Volume Expansion." *Nano Lett.* **15**(6), 3959–3963 (2015).

Zhao, Y., et al., "CH_3NH_3Cl-Assisted One-Step Solution Growth of $CH_3NH_3PbI_3$: Structure, Charge-Carrier Dynamics, and Photovoltaic Properties of Perovskite Solar Cells." *J. Phys. Chem. C* **118**(8), 9412–9418 (2014).

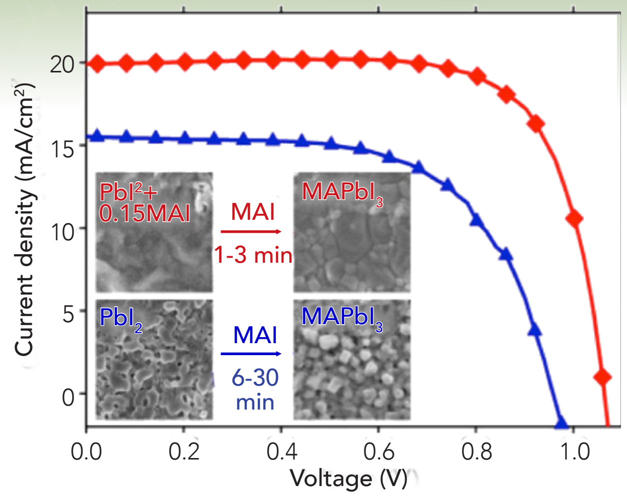
Zhao, Y., et al., "Effective Hole Extraction Using MoOx-Al Contact in Perovskite $CH_3NH_3PbI_3$ Solar Cells." *Appl. Phys. Lett.* **104**, 213906 (2014).

Zhao, Y., et al., "Efficient Planar Perovskite Solar Cells Based on 1.8 eV Band Gap $CH_3NH_3PbI_2Br$ Nanosheets via Thermal Decomposition." *J. Am. Chem. Soc.* **136**(35), 12241–12244 (2014).

Zhao, Y., et al., "Solution Chemistry Engineering Toward High-Efficiency Perovskite Solar Cells." *J. Phys. Chem. Lett.* **5**(23), 4175–4186 (2014).

Acknowledgments

The project was led by National Renewable Energy Laboratory scientists Kai Zhu, Joseph Luther, Mengjin Yang, Alexandre Nardes, and Yixin Zhao.



Demonstration of morphology control, faster phase formation, and increased efficiency by controlling the precursor chemistry of the perovskite films (T. Zhang, et al., 2015)

"Researchers demonstrated a new two-step process that speeds up the formation of the perovskite film by a factor of 10."

Creating Optimal Fracture Networks for Energy

Lawrence Livermore National Laboratory

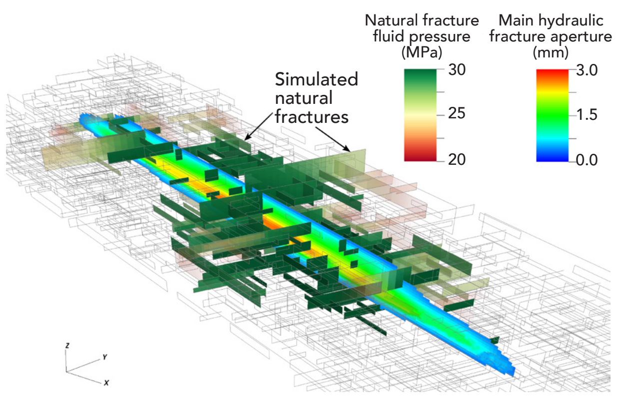
High-performance simulations supporting enhanced energy extraction with reduced environmental impact

Project Overview

A Lawrence Livermore National Laboratory project developed a computational framework, GEOS, designed to simulate hydraulic fracturing and associated fluid transport in low-permeability hydrocarbon and geothermal reservoirs. The primary goal of this project was to establish validated and extendable simulations that can be used as both an interpretive framework and predictive tool applicable to the broad range of subsurface science and engineering activities required to support energy and national security.

Why It Matters

The combination of horizontal drilling and hydraulic stimulation has revolutionized domestic hydrocarbon production. The United States is now the global leader in oil and gas production. In spite of this development, challenges remain. A large fraction of the available oil remains in the ground, and there are mounting concerns regarding water use, potential aquifer contamination, and induced seismicity from hydraulic fracturing. Enhancing the efficiency of field operations through simulation can aid in minimizing many of these concerns and result in the drilling of fewer and more efficient wells. Efficiency will not only minimize environmental impact but will also allow more of our nation's energy assets to be recovered, generating more jobs, and protecting U.S. oil companies from downward oil price pressures. "Similarly, enhancing our understanding of how fracture networks are created also benefits our comprehension of how fractures can be prevented, a critical element in minimizing the risk associated with geologic storage of carbon dioxide, radioactive waste disposal, and reinjection of water produced during hydrocarbon production," said lead investigator, geologist Rick Ryerson.



GEOS simulation of a single hydraulic fracture within a pre-existing network of natural fractures. Fracture aperture is displayed for the main hydraulic fracture and fluid pressure for the reactivated natural fractures that reveals the development of network complexity and connectivity. Natural fractures that have not been pressurized by fluid from the hydraulic fracture are rendered transparent with only perimeters shown as gray lines.

Technical & Scientific Achievements

Designed for high-performance computing platforms, GEOS simulates the growth of hydraulically driven fractures over a spatial range extending from the near-wellbore environment to the reservoir scale and realistically represents reservoir heterogeneities, stratigraphic layering, pre-existing natural fracture networks, and stress variations. GEOS also simulates the response of the fracture evolution to variations in engineering parameters. These include the depth of horizontal wells, lateral and vertical spacing of individual wells, and the location of perforation clusters. In addition, information on pumping schedules describing the temporal variations in fluid volume and viscosity, proppant fraction, and injection pressure are also incorporated. With this information, GEOS can predict lateral and vertical fracture growth, borehole pressures within the reservoir, fluid and proppant distribution, and fracture connectivity, as well as geophysical signatures such as microseismicity and surface deformation.

The results of GEOS simulations compare favorably with experimental studies, but consistency with field observations provides the ultimate measure of code validation, incorporating the influence of all the relevant, coupled physical processes and the inherent uncertainty in subsurface characterization. To access reservoir data and oil industry expertise, Lawrence Livermore has established a number of CRADAs (cooperative research and development agreements) with oil field service companies and both small independent and major oil producers. Reservoir geologic models are optimized by establishment of an initial site model followed by iterative comparison of GEOS-generated predictions of geophysical and engineering signatures with those obtained in the field. Simulations of lateral and vertical fracture growth and borehole pressure variations are in good agreement with reservoir microseimic data and downhole pressure variations, respectively.

Contributions & Impact

Given the variety of subsurface environments of interest and the coupled, multiphysics nature of hydraulic stimulation, reservoir production models are non-unique and must be tested in various settings using multiple observational methods. Incorporation of new and different observations can improve model precision and the GEOS project continues to add new capabilities such as utilization of data from fiber-optic acoustic and temperature sensors and evolving 4D seismic methods. Efforts to simulate hydrocarbon production are also being addressed by inclusion of multi-phase flow capability to simulate the transport of oil, gas, and water between the intact rock matrix adjacent to a fracture and the fracture network.

Publications

Annavarapu, C., et al., "A Weighted Nitsche Stabilized Method for Small-Sliding Contact on Frictional Surfaces." *Comput. Meth. Appl. Mech. Eng.* **238**, 763-781 (2015)

Fu, P., S. M. Johnson, and C. R. Carrigan, "An Explicitly Coupled Hydro-Geomechanical Model for Simulating Hydraulic Fracturing in Arbitrary Discrete Fracture Networks." *Int. J. Numer. Anal. Meth. Geomech.* **37**, 2278–2300 (2013).

Fu, P., S. M. Johnson, and C. Carrigan, "Using Fully Coupled Hydro-Geomechanical Numerical Test Bed to Study Reservoir Stimulation with Low Hydraulic Pressure." *Stanford Geothermal Workshop*, Stanford University Press, Palo Alto, California (2012).

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Acknowledgments

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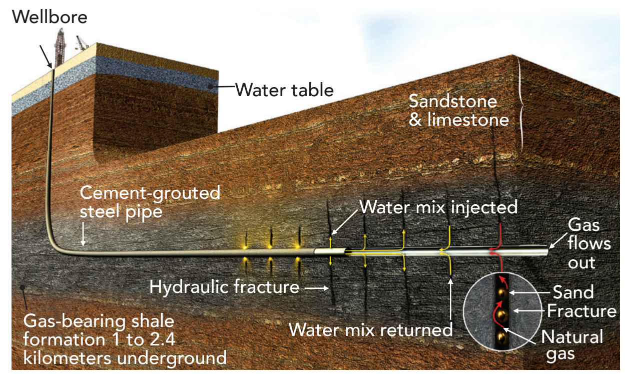


Oil and gas production from shale and other low-permeability rocks requires hydraulic stimulation, producing vertically oriented fractures that emanate from the horizontal wellbore. Proppants, typically sand or ceramic spheres, are injected to hold fractures open (inset).

"Designed for high-performance computing platforms, GEOS simulates the growth of hydraulically driven fractures over a spatial range extending from the near-wellbore environment to the reservoir scale."

Improved Forecasting Enables a More Reliable, Efficient Electric Grid

Pacific Northwest National Laboratory

Prediction software that helps regional coordinators balance energy supply and demand

Project Overview

Electric power forecasting is critical, but not easy to do. Daily, reliability coordinators at power system organizations must make quick projections for supply and demand, often without complete information. The coordinators rely on personal experience, preferred models, and past trends to predict future energy requirements, but weather and other variables increase inaccuracies. Scientists at Pacific Northwest National Laboratory have produced a software tool that overcomes these challenges by combining individual forecasting models into an aggregated collective. This approach substantially improves forecasts, which leads to lower power costs and improved grid reliability.

Why It Matters

Power forecasting errors can generate grave consequences—from blackouts to high market costs, ultimately impacting the nation's energy and economic security. For these reasons, Pacific Northwest researchers have been working to put a useful new technology in the hands of reliability coordinators, who must regularly forecast power needs and schedule exchanges of power to and from a number of neighboring entities. The sum of these future transactions, called the net interchange schedule, is submitted by the coordinators, in advance, to the balancing authorities and wholesalers that distribute power. Fluctuations in energy demand throughout the day, season, and year, along with weather events and increased use of intermittent renewable energy from the sun and wind, all impact forecasts. The methodology developed with this project has shown that it's possible to reduce errors and bring better balance between power generation and load. Further, this capability informs future forecast-related research in this and other areas, such as renewable energy integration.

Developers consult on the innovative software tool that enables improved precision for electricity forecasts, leading to lower power costs and enhanced grid reliability.

Technical & Scientific Achievements

The research team created a tool that uses algorithms developed at Pacific Nothwest to assess up to 35 different energy forecasting models and determine which models should be combined to best predict energy needs at a given time. The tool evaluates strengths of each model based on historical trends, gives the models weighted values (or eliminates them) based on that analysis, and then combines the selected models simultaneously to obtain a more accurate energy forecast. When used together, the chosen models demonstrate power in numbers by collectively generating more accurate energy forecasts than any one model alone. The method quickly adapts to the chaotic energy landscape by recognizing probable changes and picking the best combinations of models for these scenarios.

Contributions & Impact

Tests of the new capability in an actual transmission environment have produced promising results. "Forecasts 1 to 4 hours out—the most difficult to make accurately—saw a 30 to 55% reduction in errors," said Luke Gosink, a staff scientist and project lead. Forecast error reductions could significantly benefit the U.S. power system and economy, with perhaps hundreds of millions of dollars in energy savings each year. The tool, known as the Power Model Integrator, was honored with an R&D 100 Award in 2015 as one of the year's 100 best innovations worldwide.

"The underlying framework is very adaptable, so we envision using it to create other forecasting tools for electric industry use," Gosink said. For example, the methodology was included in the DOE's Lab-Corps technology transfer program, where it generated interest for application in renewable energy integration. Beyond the power industry, other uses of the technology are being explored, including predicting chemical properties studied in computational chemistry applications and identifying particles for high-energy physics experiments.

Publications

Ferryman, T., et al., "Net Interchange Schedule Forecasting of Electric Power Exchange for RTO/ISOs." IEEE Power & Energy Society General Mtg., San Diego, California, July 22–26, 2012.

Vlachopoulou, M., et al., "An Ensemble Approach for Forecasting Net Interchange Schedule." IEEE Power & Energy Society General Mtg., Vancouver, British Columbia, July 23, 2013.

Vlachopoulou, M., et al., "Net Interchange Schedule Forecasting Using Bayesian Model Averaging." IEEE Power & Energy Society General Mtg., Denver, Colorado, July 2015.

Acknowledgments

The project work was conducted by former and current Pacific Northwest researchers Ning Zhou, Luke Gosink, Alex Venizin, Maria Vlachopoulou, Jeremiah Rounds, Trenton Pulsipher, and Ryan Hafen.

"Forecast error reductions could significantly benefit the U.S. power system and economy, with perhaps hundreds of millions of dollars in energy savings each year."

Highly Efficient Low-Cost Sorbents for Groundwater Remediation of Key Radionuclides

Savannah River National Laboratory

Modified clays that can be used to clean up radionuclide-contaminated groundwater inexpensively

Project Overview

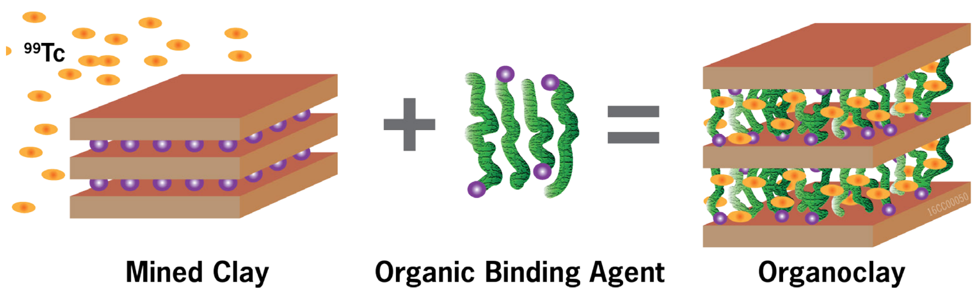
Radioiodine (^{129}I) and technetium (^{99}Tc) are key risk drivers in many DOE contaminated sites and among the top three risk drivers in active or proposed subsurface waste-disposal sites. Because of their chemical properties, they remain difficult to immobilize in the environment. The objective of this study at Savannah River National Laboratory was to develop a highly effective, low-cost sorbent that could bind with groundwater ^{129}I or ^{99}Tc, resulting in lower human and environmental risk. Two organoclays were identified that were extraordinarily effective, concentrating groundwater ^{99}Tc and ^{129}I by factors of 100,000 and 10,000, respectively. These materials cost only about $6 per kilogram and can compete in terms of efficiency with present, more costly, state-of-the-art sorbents.

Why It Matters

One of the biggest challenges facing DOE is the environmental cleanup after more than half a century of nuclear weapons production and rapidly increasing inventories of radioactive wastes generated from nuclear power production. The inventory of environmental contamination at DOE sites exceeds 6.5 trillion liters of groundwater, an amount equal to 4 times the daily U.S. water consumption, and 40 million cubic meters of soil and debris. It has been estimated that it will cost hundreds of billions of dollars and over 50 years of effort for remediation. Among the many different radionuclides in contaminated sites or in future waste disposal sites, anionic ^{129}I and ^{99}Tc pose the greatest risk, and cost-effective methods of immobilizing them continue to be a technical challenge. The high risk of these radionuclides originates from their large inventory, rapid mobility in the environment owing to their anionic nature, very long radioactive life span (half-life >200,000 years), and high toxicity (^{129}I is a thyroid seeker). Highly effective, low-cost materials for immobilizing these radionuclides are needed to reduce disposal cost and remediate contaminated groundwater and soil.

Technical & Scientific Achievements

A series of laboratory screening tests were conducted at Savannah River to evaluate sorbents for the most common and problematic anionic forms of ^{129}I and ^{99}Tc. These screening tests identified two especially effective organoclay sorbents. Organoclays are natural clay minerals that are modified with organic compounds (e.g., quaternary ammonium cations) to increase their sorption capacity. The natural montmorillonites do not bind either radionuclide, but once their surfaces were modified with the organic compounds, the binding capacities greatly improve by creating ^{99}Tc and ^{129}I binding environments. Organoclay technology is presently used for cationic metals, such as for heavy metal remediation at acid mine drainage sites. The innovation in this study was the identification of an organoclay useful for sequestering anionic contaminants. Additional testing confirmed that not only did the organoclays have large binding capacities for ^{129}I and ^{99}Tc, but they held onto the radionuclides strongly, minimizing the tendency for the bound ^{129}I and ^{99}Tc to desorb back into groundwater. Research scientist Anna Knox noted, "near irreversible sorption of these long-lived radionuclides is a requirement of this technology because they must be immobilized for a long time to assure reduction in environmental risk."



Clay mineral surfaces can be modified with organic compounds that create new binding sites for radioiodine and technetium (^{99}Tc).

Contributions & Impact

"This study permitted us to collect seed data that we used successfully in subsequent proposals," said Savannah River geochemist Daniel Kaplan. "Rather than proposing to develop an entirely untested sorbent, we were able to provide some preliminary data."

This LDRD project has led to a submitted patent disclosure. Additionally, results were used in proposals to secure (1) a three-year DOE Nuclear Energy University Program project to develop sorbents for the secondary waste generated from groundwater remediation at the Fukushima Site in Japan, (2) a DOE Environmental Management project to field test the organoclays in a ^{99}Tc plume at the Savannah River Site, and (3) a DOE Environmental Management international project to work on ^{99}Tc and carbon-14 contamination at the Sellafield Site in the United Kingdom.

Publications

Kaplan, D. I., et al., "Organo-Modified Clays for Removal of Aqueous Radioactive Anions." U.S. Patent-Pending 20150129504 A1, May 2015.

Li, D., et al., "Aqueous ^{99}Tc, ^{129}I and ^{137}Cs Removal from Contaminated Groundwater and Sediments Using Highly Effective Low-Cost Sorbents." *J. Environ. Radioact.* **136**, 56-63 (2014).

Acknowledgments

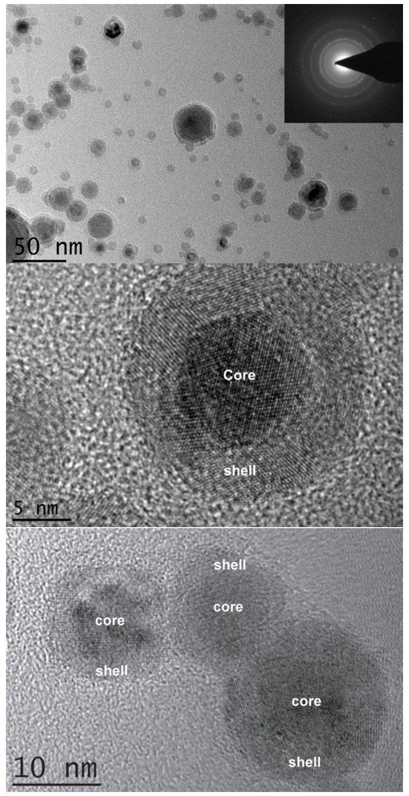
The research was led by Savannah River researchers Daniel I. Kaplan, Dien Li, Anna S. Knox, Kimberly P. Crapse, and David P. Diprete.

"Two organoclays were identified that were extraordinarily effective, concentrating groundwater ^{99}Tc and ^{129}I by factors of 100,000 and 10,000, respectively."

Magnetic Separation Nanotechnology for Spent Nuclear Fuel Recycling

Idaho National Laboratory

Understanding the usefulness of functionalized magnetic nanometer-scale particles in used nuclear-fuel separations



Bright-field transmission electron microscopy image (top) and high-resolution transmission electron microscopy images (center and bottom) show the fine structure of the core-shell iron and iron-oxide particles.

Project Overview

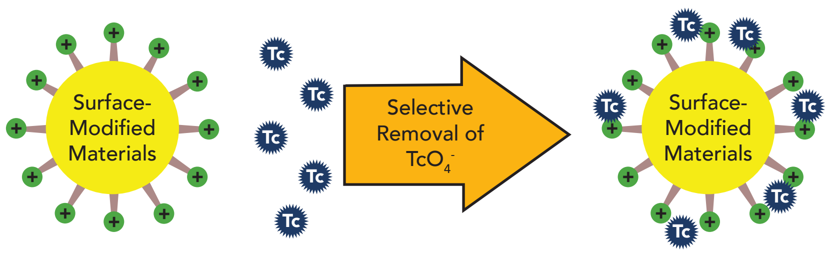
The goal of this research at Idaho National Laboratory was to develop an understanding of the usefulness of functionalized magnetic nanoparticles in used nuclear-fuel separations, particularly minor actinide/lanthanide separations and environmental remediation applications. The project helped characterize the physical and chemical properties of the ligand-functionalized magnetic nanoparticles (size, shape, aggregation, and magnetization) using electron and transmission electron microscopy, a vibrating sample magnetometer, a localized electron atom probe, and simple solid–liquid partitioning experiments with a variety of nuclides.

Why It Matters

"This study has provided promising results for application of magnetic nanoparticles materials in separations relating to used nuclear fuel separations. This will allow for safer storage options, technetium remediation, and critical material recovery from waste streams, and also contribute to the design of advanced fuel cycles and safe and effective cleanup of DOE contaminated sites," explained co-investigator You Qiang. The magnetic nanoparticle materials may also be applicable for separating rare earth minerals for extraction.

Technical & Scientific Achievements

In addition to used nuclear fuel and existing environmental waste, the research focused on the application of the magnetic nanoparticles for the remediation of technetium, which is a primary radionuclide (~6%) formed during the uranium-235 fission process. Typically found as an anionic pertechnetate species (TcO_4^-), it presents several challenges in the waste streams, as an interference in the plutonium–uranium extraction process, and poor behavior in glass vitrification. To address these issues, investigators developed a surface-modified material for the selective removal of TcO_4^- for application with magnetic nanoparticles for facile separation of technetium from an aqueous waste stream. A solid-phase material was modified to specifically extract TcO_4^- based on previous studies at Washington State University.



Selective removal of an anionic pertechnetate species (TcO_4^-) using surface-modified materials for use with magnetic nanoparticles for separation of technetium from aqueous waste streams.

The general experimental approach involved using batch-separation studies with the surface-modified material to determine the overall performance to extract $^{99m}TcO_4^-$ from aqueous solutions. The impact of acid or base on the TcO_4^- extraction behavior was also evaluated to determine the efficacy of the material over a pH range.

Because of the complex nature of wastes stored in underground tanks, competing anions can potentially limit the overall effectiveness of TcO_4^- extraction with the surface-modified material. Several common anions including chloride, sulfate, phosphate, nitrite, nitrate, and perchlorate were examined as a function of anion concentration on the TcO_4^- extraction. Future studies involving the surface-modified material will examine the impact of a reprocessing simulant solution and reprocessed tank waste to mimic conditions that would be encountered during the reprocessing streams, where multiple anions are competing simultaneously.

Contributions & Impact

The work is contributing towards the design of an advanced fuel cycle that is proliferation-resistant and enables safe and effective cleanup of DOE contaminated sites as well as cost-effective recovery of metal ions in solution from mining and ore-refining activities.

Publications

Zhang, H., et al., "Selective Extraction of Heavy and Light Lanthanides from Aqueous Solution by Advanced Magnetic Nanosorbents," *ACS Appl. Mater. Interfac.* **8**(14), 9523-9531 (2016).

Acknowledgments

This work was led by principal investigator Leigh Robert Martin of Idaho National Laboratory. Co-investigators included You Qiang from University of Idaho, Yaqiao Wu from Boise State University Center for Advanced Energy Studies, and Paul Benny from Washington State University.

Hujin Zhang, a University of Idaho Ph.D. student and intern at Idaho National Laboratory, looks at an agglomeration of magnetic nanoparticles post-extraction.

Leigh Martin performs fluorescence experiments to monitor binding of lanthanides to the magnetic nanoparticles.

"This will allow for safer storage options, technetium remediation, and critical material recovery from waste streams, and also contribute to the design of advanced fuel cycles and safe and effective cleanup of DOE contaminated sites."

Rapid Viability Assays for Biothreat Events

Pacific Northwest National Laboratory

Rapid, integrated approach to determine the viability of disease-causing bacteria

Project Overview

When a potential biothreat such as *Bacillus anthracis*, *Salmonella* spp., or *Escherichia coli* arises, it's crucial to quickly identify the biological material and understand the nature of the threat. A key question is whether the organism is alive and thus can cause disease. Researchers at Pacific Northwest National Laboratory developed a method, in conjunction with a smartphone-based microscope, which can be used in the field to quickly and efficiently help determine the course of action when facing an unknown biothreat.

Why It Matters

Like an old seed that can never germinate, biological agents that are not alive (viable) can't grow or cause disease. Currently, scientists must cultivate the unknown biological material in a laboratory to determine identity and show whether it is alive or dead. This approach typically takes many hours or even days, thus delaying crucial decisions for a biothreat response, remediation, and recovery. In this project, investigators developed new methods to inform both response and recovery for field-based detection to enable first responders and scientists.

Technical & Scientific Achievements

Researchers at Pacific Northwest developed and optimized a rapid growth and optical detection method for the anthrax surrogate known as *Bacillus anthracis* Sterne. A specialized sample holder, or a microfluidic incubation device, is used to germinate and view the spores of the anthrax surrogate. The spores are allowed to grow and form strands of bacteria that are easily viewed using smartphone microscopy. Stained with fluorescent dyes, live cells capable of causing disease show as green strands. Dead cells, which can't cause disease, show as yellow, orange, or red strands.

The microfluidic sample holder works even when there are low numbers of bacteria, has built-in filters, lets users position and concentrate the bacteria for viewing, and enables a secondary confirmation to validate the results.

A transformational and unexpected outcome of this project was the development of a 3D printable attachment for a smartphone, creating a portable microscope. The microscope consists of a 3D-printed housing that supports a glass bead that functions as the microscope lens. Other existing smartphone microscopes are bulky, expensive, hard to align, or are lower powered. The smartphone microscope can magnify a sample by 750 times to visualize tiny pathogens, and the slip-on plastic housing can be thrown away if contaminated. It costs pennies to make one, as compared with a typical cost of around $3,000 for a microscope with similar capabilities in a laboratory setting.

Contributions & Impact

This LDRD project has reduced overall detection time by fourfold. The reduction of time to accurately detect and respond to a natural or human-caused biothreat translates directly into number of lives saved. Conversely, if the biological material is determined to be inactive, officials need not launch a remediation and recovery effort.

An additional bonus is the impact and widespread popularity that the printable 3D microscope is having in science, technology, engineering, and math education, creating hands-on experiences. The design specifications are available online from Pacific Northwest, free of charge, so anyone with access to a 3D printer can make their own microscope.

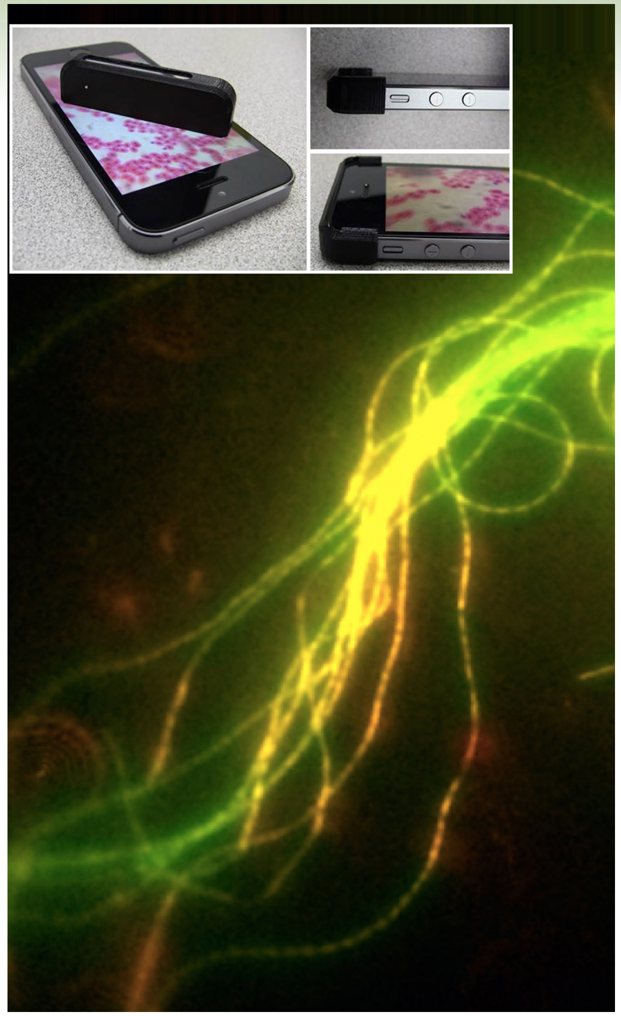
Publications

Hutchison, J. R., and R. L. Erikson, "Detecting Anthrax in the Palm of Your Hand: Applications of a Smartphone Microscope." *Atlas of Science* (November 14, 2015).

Hutchison, J. R., et al., "Reagent-Free and Portable Detection of *Bacillus anthracis* Spores Using a Microfluidic Incubator and Smartphone Microscope." *Analyst* **140**(18), 6269–6276 (2015).

Acknowledgments

This work is a multidisciplinary collaboration of Pacific Northwest scientists and engineers including Janine Hutchison, Rebecca Erikson, Brooke Deatherage Kaiser, Rich Ozanich, Becky Hess, Josef Christ, and Ryan Kelly.



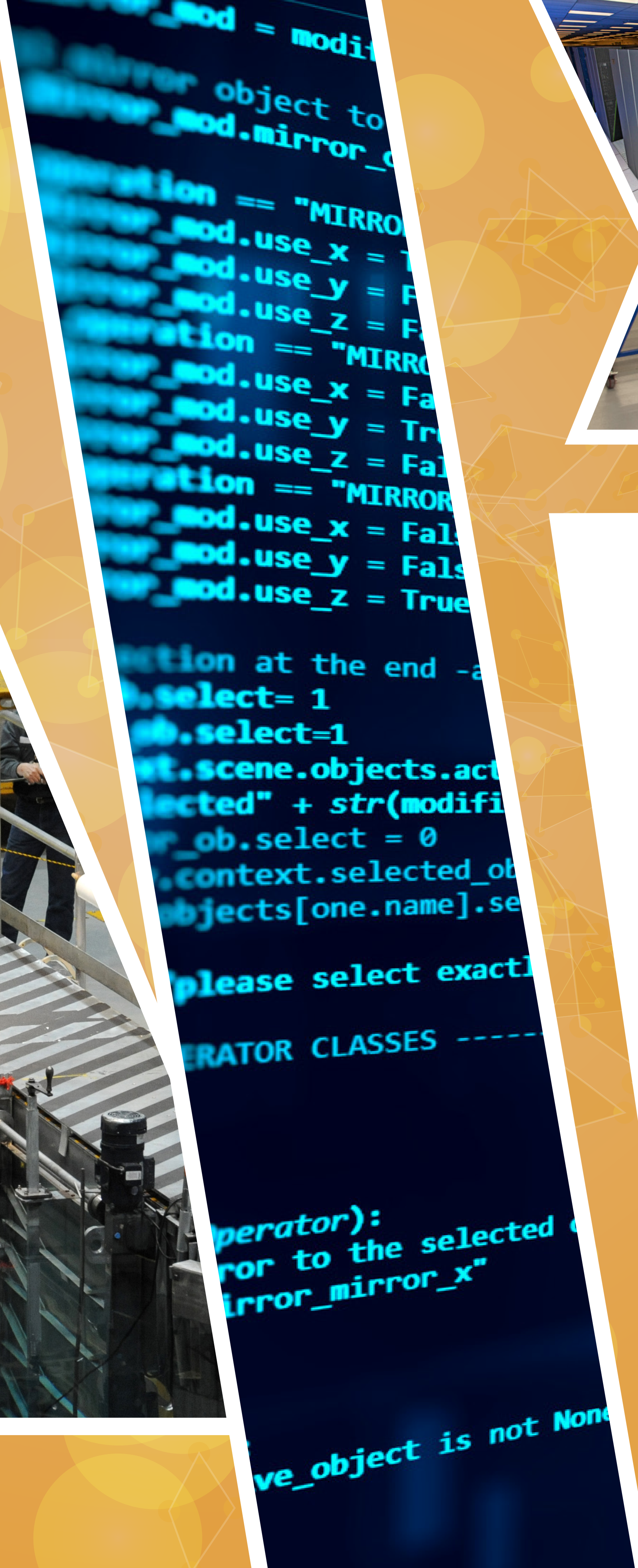
Using the low-cost smartphone microscope developed with this project, potential biothreats like this surrogate bacteria for the disease anthrax can be visualized and assessed quickly and easily in the field.

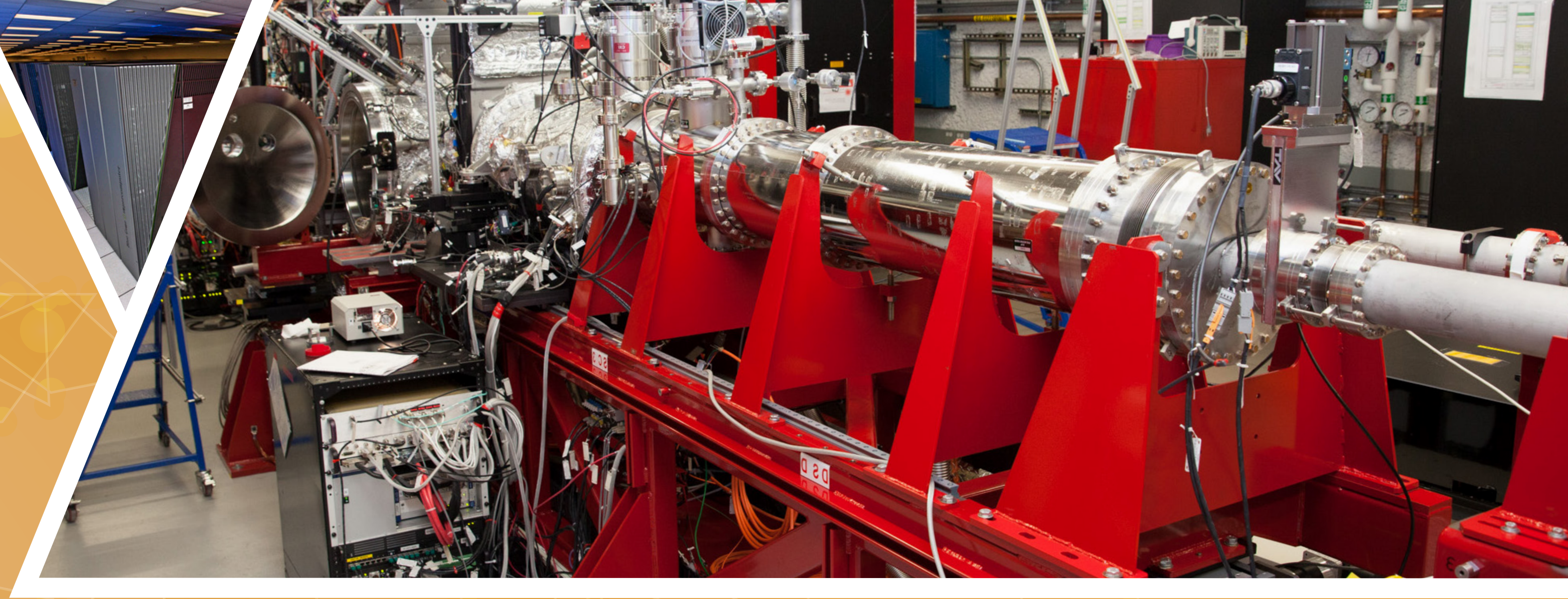
"The reduction of time to accurately detect and respond to a natural or human-caused biothreat translates directly into number of lives saved."

NUCLEAR SECURITY

From the program's inception, a priority of LDRD has been to advance the frontier of basic science and technology to enhance nuclear security through defense, nonproliferation, forensics, and remediation efforts. LDRD projects have made essential contributions to every facet of nuclear security, including stockpile stewardship, high-energy-density research, high-performance computing and simulation, and nuclear and isotopic science. For the stockpile stewardship mission, for example, LDRD researchers developed unique capabilities to measure microstructural, electrical, and chemical properties of plutonium and its alloys used as the core, or pit, of a nuclear weapon. A decade of sustained LDRD investments examined the effect of decay on crystal structure and resultant changes in material properties, laying the foundations for the weapons laboratories to create "accelerated" aged alloys and thus study the equivalent of 60-year-old plutonium in just 4 years. The most accurate science-based estimates ever obtained for pit lifetimes were derived by combining experimental and computational resources. These LDRD and subsequent DOE investments resulted in eliminating the need for a proposed large and costly Modern Pit Facility.

In addressing emerging nuclear security needs, LDRD continues to develop impactful technologies, including proton radiography for imaging materials under shock compression and special materials and processing techniques to make radiation-hardened microchips for use in weapons. LDRD is also addressing gaps in existing technologies to detect controlled radiological materials that could be smuggled in with cargo at U.S. ports with a new plastic scintillator that responds faster to telltale emissions and discriminates threat materials from benign radiation sources. An LDRD-developed power spectrum analysis device is being used to determine how aging processes affect device behavior in non-nuclear weapons components. LDRD is also creating new manufacturing technologies for materials used as structural weapon components that are "self-sensing" and can report on their health status. In addition, basic LDRD research to understand the mysteries of cosmic explosions is providing insight into the physics of nuclear explosions with an automated software system created for supernovae studies.





NUCLEAR SECURITY

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30 Spectral "Fingerprints" Give Clues about Device Behavior

32 Multifunctional Materials and Manufacturing

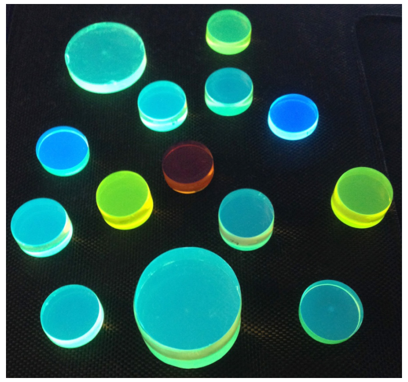
34 Supernovae for National Security

36 Revolutionizing Nuclear Forensics with Laser-Induced Breakdown Spectroscopy

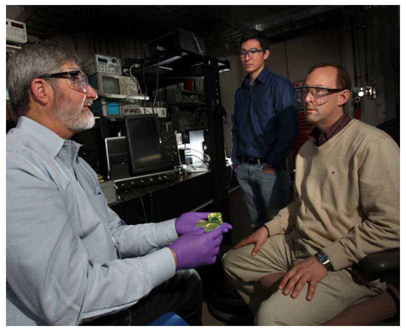
Lighting Up the Science of Radiation Detection

Sandia National Laboratories

New technology providing an effective, less-costly means of identifying nuclear materials



Triplet-harvesting plastic scintillators under 365-nanometer illumination.



Sandia researchers Patrick Doty, Patrick Feng, and Mark Allendorf (L to R) have created a new type of scintillator using metal organic framework or plastic scintillator hosts combined with heavy metal dopants, shown in Doty's hand. This material enables detection of neutrons and gamma rays using spectral discrimination techniques that could transform radiation detection. (Photo by Dino Vournas.)

Project Overview

Millions of barrels of cargo are unloaded from ships to U.S. soil every year. Automated sensors screen cargo at ports of entry for controlled radiological materials that could be used to make a nuclear weapon. The detectors scintillate (glow) when they pick up telltale emissions. Sandia National Laboratories researchers have developed a new plastic scintillator that gives off more light at less cost, and responds faster than current scintillators. The unique timing response also provides the ability to discriminate threat materials from benign radiation sources.

Why It Matters

Federal agencies continuously seek new radiation detection technologies for special nuclear materials identification and treaty verification. These scintillators promise to revolutionize special nuclear material detection in challenging, real-world scenarios such as active screening of cargo containers. Triplet-harvesting plastic scintillator (THPS) technology housed inside radiation portal monitors could quickly screen cargo for controlled radiological materials, even in environments where benign background radiation is high.

"Improving our radiation detection capabilities is crucial to advancing NNSA's nonproliferation mission," said Anne Harrington, NNSA's deputy administrator for Defense Nuclear Nonproliferation. "Preventing the illicit movement of radiological and nuclear materials around the globe supports. . .nuclear security objectives and helps to mitigate the threat of a nuclear terror attack."

Technical & Scientific Achievements

Fundamentally different from existing technology, the THPS is composed of heavy-metal dopants—a trace impurity that alters the optical properties—housed in a host polymer. The respective energy levels of the polymer and dopant compounds are tuned such that energy transfer is possible. Upon interaction with ionizing radiation, the polymer matrix is left in an excited electronic state, which then relaxes to emit light. The triplet harvesting process, which refers to the spin multiplicity of the excited electronic state, takes advantage of a phenomenon known as spin–orbit coupling. In this mechanism, the triplet-harvesting additive "scavenges" the excited-state energy not converted to light, resulting in a bright, colored glow.

Triplet harvesting controls the relative brightness, color, and timing characteristics of the respective emissions. The proportion of host-to-dopant luminescence is specific to a particular type of radiation (gamma or neutron). In particular, uncharged fast neutrons and gamma rays interact with THPS materials to produce recoil protons and scattered electrons, respectively. These charged particles are absorbed within the material and lead to distinct emission properties. Using spectral-shape discrimination, gamma and neutron particles can be distinguished from one another based on the color of the emitted light: fast neutrons produce a larger ratio of green to blue luminescence. Conventional pulse-shape discrimination techniques, which utilize timing differences, can also be used with the THPS. Thus, this technology could be used as an upgrade to non-discriminating plastic scintillator systems, as a "drop-in" replacement for liquid-based systems, or as part of a simplified optical configuration using spectral-shape discrimination.

This 2014 R&D100 award-winning technology is rooted in two LDRD projects. From 2005–2008, principal investigator Mark Allendorf utilized metal–organic framework materials to study various phenomena, principally comprising luminescence and scintillation properties. In 2010, Patrick Doty set out to determine if it was possible to convert non-luminescent energy states from a host material (metal–organic framework, liquid, or polymer) to a heavy-metal dopant compound. Doty used the knowledge gained from Allendorf's project to successfully demonstrate the concept using metal–organic framework materials as a "nano-laboratory." At the conclusion of this LDRD project, post-doctoral researcher Patrick Feng (now staff), working with both Doty and Allendorf, proposed to extend the research to polymer materials. Feng's team won funding from NNSA to develop the THPS concept.

Contributions & Impact

This LDRD-based capability addresses critical shortcomings of existing technologies used for fast neutron detection and discrimination. Compared to existing materials, the new scintillators offer a 30% improvement in brightness, significantly faster timing response, and unprecedented optical discrimination capabilities. The cost is also nearly 10 times less than the closest competing material. This combination of features enables THPS materials to operate under the low-signal, high-background, and high-rate environments encountered in demanding real-world scenarios. THPS materials enable the extraction of vital diagnostic information from mixed radiation fields—information that has remained elusive until now.

Publications

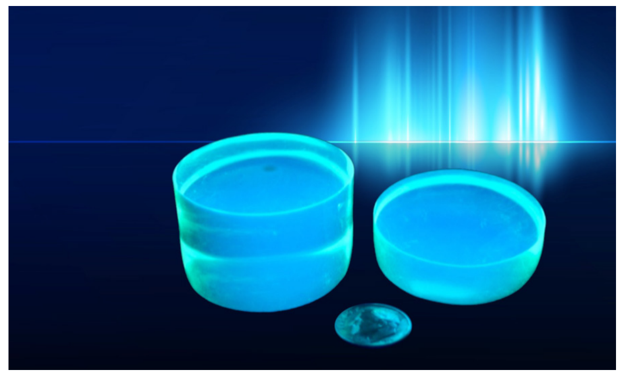
Doty, F. P., M. D. Allendorf, and P. L. Feng, "Doped Luminescent Materials and Particle Discrimination Using Same." U.S. Patent 20110108738 A1, May 2011.

Feng, P. L., and M. E. Foster, "Pulse-Shape Discrimination in High-Symmetry Organic Scintillators." *IEEE Trans. Nucl. Sci.* **60**(4), 3142–3149 (2013).

Feng, P. L., et al., "Designing Metal-Organic Frameworks for Radiation Detection." *Nucl. Inst. Meth. Phys. Res. A* **652**(1), 295–298 (2011).

Acknowledgments

Initial work funded by Sandia's LDRD program was led by Mark D. Allendorf and F. Patrick Doty, with key contributions from Patrick L. Feng.



Triplet-harvesting plastic scintillators under 365-nanometer illumination. (Dime shown for scale.)

"These scintillators promise to revolutionize special nuclear material detection in challenging real-world scenarios."

Spectral "Fingerprints" Give Clues About Device Behavior

Sandia National Laboratories

Investigating aging effects in weapons components with power spectrum analysis

Project Overview

Researchers at Sandia National Laboratories are using power spectrum analysis (PSA) to investigate aging effects in non-nuclear weapons components. PSA is a technique that measures a unique device signature when the device is subjected to a stimulus. This signature is like a fingerprint. By comparing the "fingerprint" of an un-aged device to that of an aged device, researchers can gain insight into early detection and how aging processes might affect device behavior. For some devices, PSA detected aging effects at greater sensitivity than conventional methods, showing promise as a tool for stockpile assurance.

Why It Matters

The long-term reliability and lifetime of nuclear weapons components are major concerns of an aging stockpile. The ability to predict the effects of aging early on and detect potential failures would be invaluable in maintaining the integrity and reliability of the stockpile.

Technical & Scientific Achievements

NNSA's stockpile stewardship mission is aimed at ensuring the safety, security, and reliability of weapons in the absence of underground nuclear tests. Understanding how new and existing weapons components will behave throughout the life of the system is critical to maintaining the stockpile. By performing accelerated-aging tests on components, researchers can observe device behavior. In the absence of obvious degradation, other techniques must be used to detect, understand, and predict aging effects. Researchers are using Sandia-developed PSA to detect electrical differences in devices and determine whether PSA can detect aging effects when devices—such as commercial-off-the-shelf discrete devices, diodes, and capacitors—are subjected to accelerated life tests at elevated temperatures and voltages.

After each accelerated life-test cycle, researchers made PSA measurements and performed conventional electrical tests on the devices. They also performed physical analyses on several aged and un-aged devices. By correlating PSA data and the conventional data, they observed promising results. PSA showed good sensitivity to detecting aging effects in certain types of devices. In these cases, the aging effects detected were not identified by conventional electrical means. Additionally, they found strong correlations between changes in PSA data and physical changes in the test devices. These initial results suggest that PSA can potentially be used to study aging effects as a stand-alone technique or as a complementary technique to existing electrical testing methods, providing a useful tool for stockpile assurance.

LDRD has been critical in the development and application of PSA by helping to develop a fundamental understanding of the physical mechanisms that generate PSA signatures, providing insight into the nonlinear processes that generate PSA signatures, as well as a better understanding of the limitations and detection sensitivity of PSA. Developed initially for counterfeit detection, this nondestructive, efficient technique has been shown to be a highly effective screening tool.

Contributions & Impact

The PSA technique is being considered for use in device screening for high-consequence nuclear security applications. The method has successfully detected counterfeit devices, differences between counterfeit and genuine parts, and differences in manufacturers, foundries, and features. The method has also detected aged devices, irradiated parts, and changes to processing and packaging. In November 2015, a patent was awarded for defect screening in integrated circuit devices.

Publications

Tangyunyong, P., and G. Lourbriel, "Power Spectrum Analysis." Sandia National Laboratories, SAND2016-6767PE (2016).

Tangyunyong, P., E. Cole, Jr., and D. Stein, "Power Spectrum Analysis for Defect Screening in Integrated Circuit Devices." U.S. Patent 9188622 B2, November 2015.

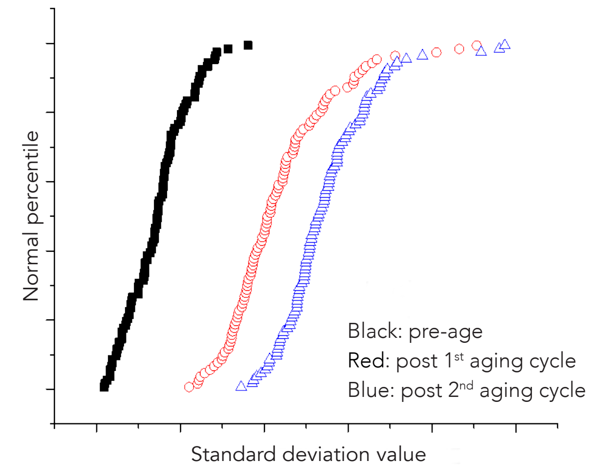
Tangyunyong, P., et al., "Aging Detection in Zener Diodes Using Power Spectrum Analysis (PSA)." GOMACTech 2016, Orlando, Florida, March 2016.

Tangyunyong, P., et al., "Counterfeit Detection Using Power Spectrum Analysis (PSA)." GOMACTech 2015, St. Louis, Missouri, March 2015.

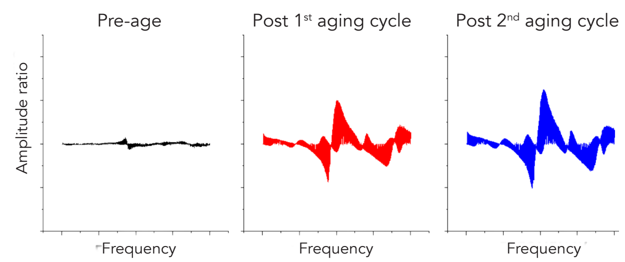
Acknowledgments

PSA was enabled by Sandia's LDRD, Enhanced Surveillance, and Component Maturation Development programs. The following LDRD projects were led by Sandia researcher Paiboon Tangyunyong, and include the researchers listed below:

- Biliana Paskaleva, Rachelle Thompson, Ed Cole, and Darlene Udoni contributed to project 180864 ("Predictive Assessment of State of Health and Life Time of Components").
- Larry Bacon, Josh Beutler, Mark Jenkins, Eric Keiter, Alan Mar, Thomas Russo, and Ralph Young contributed to project 171057 ("Comparative Approach for a Physics-Based Understanding of Power Spectrum Analysis Signatures").
- Gad Hasse and Kevin Fox contributed to project 176309 ("Effects of PSA Measurements on Device Reliability") .
- Ed Cole and Kevin Fox contributed to project 154273 ("Beam-Enhanced Electrical 'Fingerprinting' of Complex Integrated Circuits").



Distributions of test diodes before aging and after two successive aging cycles. Most of the aging occurs after the first aging cycle.



Power spectrum analysis signatures of a representative diode before aging and after two successive aging cycles.

"The ability to predict the effects of aging early on and detect potential failures would be invaluable in maintaining the integrity and reliability of the stockpile."

Multifunctional Materials and Manufacturing

Lawrence Livermore National Laboratory

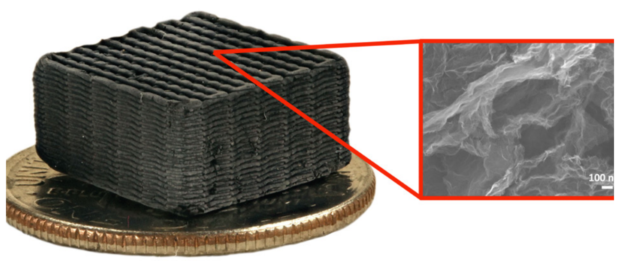
New manufacturing technologies and design methods that lead to materials and structures with multiple functionalities

Project Overview

Additive manufacturing—a breakthrough technology that is revolutionizing domestic and global manufacturing—creates the potential to engineer materials possessing desired structural, thermal, electrical, chemical, and photonic properties in a single construct. The goal of this project at Lawrence Livermore National Laboratory was to develop new design methodologies and manufacturing techniques that will enable production of specific multifunctional materials with mission-relevant applications ranging from stockpile stewardship to global security.

Why It Matters

The applications of multifunctional materials having microscale to nanometer-scale features and composed of mixtures of materials are broad and impactful. Therefore the ability to design and fabricate them is important. Some examples include electrochemical systems such as batteries and supercapacitors that are capable of carrying a mechanical load, as well as structural components with electrical pathways and integrated sensing resulting in "self-sensing" materials that can report out on their health status. These types of materials are having broad use throughout Lawrence Livermore's Stockpile Stewardship and Global Security programs.



3D printed graphene aerogel lattice with nanometer-scale features for use as a high-performance structural electrode in supercapacitors.

Technical & Scientific Achievements

Fabricating such revolutionary multifunctional materials requires new fabrication methods, typically based on additive manufacturing. Researchers have pursued several innovative new techniques including

- Direct Metal Writing: Heating metal alloys to form semi-solid pastes that can then be extruded through fine nozzles.
- Diode-Based Additive Manufacturing: Utilizing high-power laser diode arrays to melt entire layers of metal powder in a single light exposure.
- 3D Multibeam Lithography: Projecting 3D holographic light patterns into photo-curable liquid resins to create entire 3D structures in a single exposure.
- Large-Scale Optical Trapping: Allowing for the manipulation and assembly of microscale and even nanometer-scale particles.

Bench-top prototype systems functioning for all of these concepts are now available, and have been used to create structures and materials of interest.

Additionally, feedstock materials development is a critical aspect of this work. "In one of the best examples of a multifunctional material, our team has been able to 3D print graphene aerogel into designed lattice structures," relates principal investigator, engineer Chrsitopher Spadaccini. In these structures, there are mesoscale features as a result of the printing technology. However, at the microscale and nanometer-scale, graphene platelets can be observed. Graphene is an ultrahigh-surface-area material that has excellent chemical and electrical properties, ideal for batteries and supercapacitors. This work is combining the structural properties of the lattice architecture at the mesoscale with the electrochemical functionality of the graphene at the nanometer-scale to form a multifunctional material—a high-performance supercapacitor that is strong and compressible. The figure on the previous page shows the printed material with a magnified view of the graphene at the microscale. The printed supercapacitor has excellent charge and discharge performance, high energy and power density, is extremely lightweight, and can be compressed to 95% strain and fully recover to its original form. This type of material is ideal for lightweight electronics applications as well as power systems that fit into small volumes.

Contributions & Impact

This work has been impactful for both the Lawrence Livermore Stockpile Stewardship and Global Security programs. Specifically, some of the fabrication technologies and materials that have been developed are being adopted by important programmatic application teams, including those working in energy storage, carbon dioxide capture, and specific national security areas. It has also garnered significant commercial interest through our patent portfolio with several patents licensed and multiple-industry CRADAs (cooperative research and development agreements) moving forward. Finally, the work has garnered significant attention in the academic community as evidenced by multiple high-impact publications.

Publications

Hopkins, J. B., et al., "Polytope Sector-Based Synthesis and Analysis of Microarchitectured Materials with Tunable Thermal Conductivity and Expansion." *J. Mech. Des.* **138**(5), 051401–051401-10 (2016).

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Zhu, C., et al., "Highly Compressible 3D Periodic Graphene Aerogel Microlattices." *Nat. Comm.* **6**, 6962 (2015).

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Acknowledgments

This work included a large team of Lawrence Livermore staff, post-doctoral associates, and students. The project was led by Livermore researchers Chris Spadaccini, Joshua Kuntz, Eric Duoss, Andrew Pascall, and Marcus Worsley. External collaborators included Professors Nicholas Fang (Massachusetts Institute of Technology); Jennifer Lewis (Harvard University); Jonathan Hopkins (University of California, Los Angeles); Yat Li (University of California, Santa Cruz); and Daniel Tortorelli (University of Illinois at Urbana Champaign).

"Fabrication technologies and materials that have been developed are being adopted by important programmatic application teams, including those working in energy storage, carbon dioxide capture, and specific national security areas."

Supernovae for National Security

Los Alamos National Laboratory

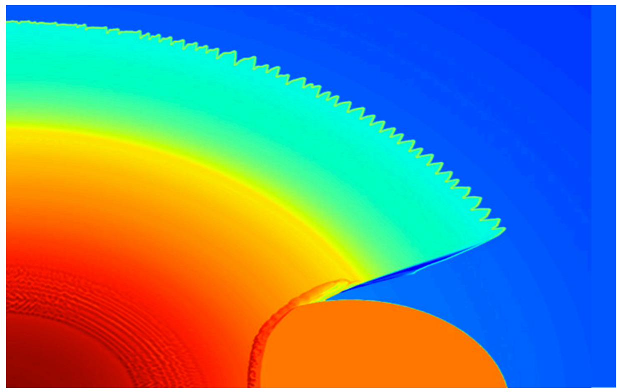
Basic research and development to understand the mysteries of cosmic explosions providing insight into the physics of nuclear explosions

Project Overview

Every computer simulation of a core-collapse supernova, in which an imploding stellar core triggers an explosion, includes code that accounts for turbulence in the core and body of the star. The only way to be sure that code is correct is to simulate the system and compare the output with what is seen in images of a real supernova. Interestingly, turbulence also shows up in another system that first implodes and then explodes—the core of a nuclear weapon. Like astrophysicists, weapons scientists need to validate their computer codes, and because the underlying physics governing the turbulence is the same between the two systems, Los Alamos National Laboratory weapons scientists have been able to better understand weapons by modeling supernova.

Why It Matters

Supernovae play an important role across a wide range of fields in physics and astronomy—they are an important source of energy to galaxies, yield exotic objects like neutron stars and black holes, and create heavy elements that make life possible. Interestingly, they also require the same numerical and physics expertise in which Los Alamos scientists have excelled.



A Los Alamos simulation of an exploding white dwarf, in which the supernova drives an expanding shock wave that collides with a torus of material accreted from a companion star.

The importance of supernovae became even more critical with the realization that Type Ia supernovae can be used to gauge cosmic distances by how dim they appear relative to their actual brightness. This enabled one of the major scientific discoveries of 1990s—measuring the accelerated expansion of the universe—and galvanized the scientific community to use supernovae to characterize dark energy.

Technical & Scientific Achievements

A robotic observing system known as the intermediate Palomar Transient Factory is a leader among the new breed of data-intensive sky monitoring surveys that seek to discover and understand transient events of astrophysical origin. Using an automated software system developed at Los Alamos that reliably identifies real astronomical transients, the survey discovered a peculiar Type Ia supernova, designated iPTF14atg. Type Ia supernovae occur in binary systems when two stars orbit one another and one of the stars is a dense white dwarf. Thanks to the software, iPTF14atg was discovered quickly enough to see what astronomers believe to be the collision between the supernova shock and its companion star, suggesting that its companion was a more-or-less normal star.

Contributions & Impact

The automated software system addresses the challenges of selecting transients from the torrent of images captured by the intermediate Palomar Transient Factory and quickly identifying the ones that need further attention. The benefit is multi-pronged—the machine learning algorithms used to automate and optimize the entire process of selecting, vetting, and prioritizing transients can also be applied to other "needle in a haystack" problems like nuclear nonproliferation.

Another LDRD project utilized Los Alamos supercomputing capabilities and a high-performance computational fluid dynamics code to perform the largest turbulent reacting flow simulations of Type Ia supernovae to date. How the cosmic explosion of a Type Ia supernova occurs is not fully understood. For example, the debate around burn rate and explosion mechanics is still not settled. In addition, the flame speed—that is the rate of expansion of a flame front in a combustion reaction—remains a big unknown. While solving the flow problem in a whole supernova is still far in the future, accurately solving the turbulent flow in a small domain around a single flame (characterizing the early stages of the supernova) has become possible. The very high-resolution reacting turbulence simulations enabled by the Los Alamos supercomputers can probe parameter values close to the detonation regime, where the flame becomes supersonic, and explore for the first time the turbulence properties under such complex conditions.

Supernovae research is a clear example of how basic science can support the weapons program. It can be quite costly to obtain data that can be used to test weapons codes, but astronomers generate new supernova data every day. With Los Alamos researchers having ties to both the weapons program and the international astrophysics community, software developed for supernovae observations can benefit weapons physics. In fact, algorithms developed by these LDRD projects have driven novel approaches for computational physics in NNSA's Advanced Simulation and Computing program. An added benefit of this work is the recruitment of new staff into the weapons physics program. In fact, 7 of the 10 postdoctoral researchers who contributed to this project were converted to staff and continue to work in applied programs at Los Alamos.

Publications

Cao, Y., et al., "Ultraviolet Radiation from Supernova-Companion Collision in a Type Ia Supernova." *Nature* **521**(7552), 328 (2015).

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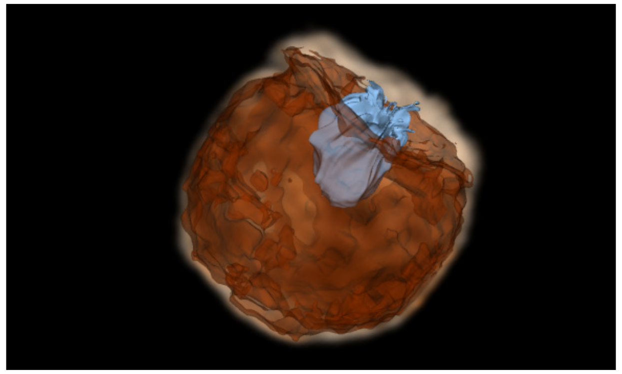
Acknowledgments

Chris Fryer, Przemyslaw Wozniak, and Daniel Livescu led the LDRD projects that supported this work, with contributions from other other researchers at Los Alamos and many collaborators at DOE laboratories, universities, and several national and international institutes.



In this still from a simulation, a Type Ia supernova explodes (dark brown color). The supernova material is ejected outwards at a velocity of about 10,000 kilometers per second. The ejected material then crashes into its companion star (light blue color). The violent collision produces an ultraviolet pulse that is emitted from the conical hole carved out by the companion star. (Image courtesy of Daniel Kasen.)

"Supernovae research is a clear example of how basic science can support the weapons program."

Revolutionizing Nuclear Forensics with Laser-Induced Breakdown Spectroscopy

Los Alamos
National Laboratory

Elemental analysis with laser-induced breakdown spectroscopy (LIBS) that pays off in space and on the ground

Project Overview

Laser-induced breakdown spectroscopy (LIBS) is a technique largely developed at Los Alamos National Laboratory to determine the composition and concentration of elements in a variety of sample types. LDRD has made many strategic investments in LIBS in the last 15 years, enabling researchers to develop the technology for diverse applications, from treaty verification to space exploration.

Why It Matters

One of the primary motivations for working on LIBS is to provide tools that can be used by inspectors to rapidly detect the approved or unapproved use of nuclear material. In most cases, LIBS can assay nuclear or nuclear-related materials in a matter of minutes, with little-to-no sample preparation. More traditional analysis methods can take days or longer for the same determination, and often require sample preparation that takes much longer than the actual measurement.

Technical & Scientific Achievements

Inspired by International Atomic Energy Agency needs, LDRD researchers at Los Alamos miniaturized the LIBS equipment for atomic emission analysis so that it can be carried on an operator's back. The Backpack LIBS system weighs only 25 pounds, is ergonomically designed, and is easy to use. Moreover, the safety features of Backpack LIBS ensure that an operator is free from laser-beam exposure and doesn't even need to wear safety glasses. Thanks to its portability, accuracy, and cost effectiveness, Backpack LIBS inexpensively takes atomic emission analysis from a traditional laboratory setting into the field. Now inspectors can use the LIBS Backpack system as a screening tool to locate special nuclear material either outside or inside facilities.

Contributions & Impact

Los Alamos uses LIBS to address non-nuclear challenges as well. An LDRD project explored new modes of analysis for LIBS that were later applied to ChemCam, a rock-blasting laser on the NASA Mars rover, Curiosity. ChemCam can detect any element on the periodic table by zapping anywhere within about 23 feet of the rover vehicle. Recent findings published in the journal *Science* showed substantial bodies of water likely existed on the surface of Mars in its early history, including long-lasting lakes that built up deposits at least 250-feet deep, and likely much deeper. More recently, ChemCam observed high levels of manganese oxides in Martian rocks, suggesting that higher levels of atmospheric oxygen once existed on Mars. This hint of more oxygen in Mars' early atmosphere adds to other Curiosity findings revealing how Earth-like our neighboring planet once was. Because of ChemCam's success, an updated version has been selected for the Mars 2020 mission as well.

In yet one more application of the technology, Los Alamos researchers developed a version of the LIBS Backpack system specifically for the oil industry. Throughout the world, oil, gas, and petrochemical plants use vessels and pipes to store or transport fluids. Over time, some of them can corrode because of the caustic nature of the fluids inside them. Pipe LIBS identifies their elemental composition in a matter of seconds, making it possible to inspect pipes, vessels, and other components already operational in the field, as well as new ones that are received from manufacturers.

Like many LDRD investments, LIBS was initiated with one mission in mind and ended up opening doors for several other seemingly unrelated missions. Investment in the research and development of LIBS has resulted in new instruments and techniques for national security, energy, space, and commercial applications, and Los Alamos researchers aren't finished yet!

Publications

Clegg, S. M., et al., "Planetary Geochemical Investigations Using Raman and Laser-Induced Breakdown Spectroscopy." *Appl. Spectros.* **68**(9), 925–936 (2014).

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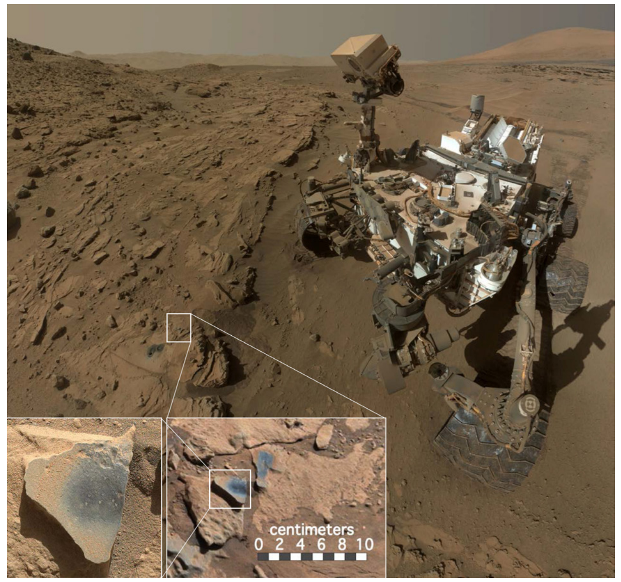
Wiens, R. C., et al., "Joint Analyses by Laser-Induced Breakdown Spectroscopy (LIBS) and Raman Spectroscopy at Stand-Off Distances." *Spectrochim. Acta Mol. Biomol. Spectros.* **61**(10), 2324–2334 (2005).

Acknowledgments

Samuel Clegg and Roger Wiens led the LDRD projects that supported this work, with contributions from other researchers at Los Alamos National Laboratory, and collaborators at Oak Ridge National Laboratory; Idaho National Laboratory; Argonne National Laboratory; Lawrence Berkeley National Laboratory; University of Nevada at Las Vegas; Pennsylvania State University; University of New Mexico; New Mexico State University; University of California, Berkeley; and several other national and international institutes.



The Curiosity rover examines the Kimberley formation in Gale Crater, Mars. In front of the rover are several flat, dark-toned features that have been cleared of dust (see inset images). These flat features are erosion-resistant fracture fills composed of manganese oxides, which require abundant liquid water and strongly oxidizing conditions to form. The discovery of these materials suggests that the Martian atmosphere might once have contained higher abundances of free oxygen than in the present day. (Image credit: MSSS/JPL/NASA, PIA18390.)

"Like many LDRD investments, LIBS was initiated with one mission in mind and ended up opening doors for several other seemingly unrelated missions."

SCIENTIFIC INNOVATION & DISCOVERY

LDRD is a major vehicle of scientific innovation vital to the national laboratories, allowing scientists to respond to national priorities, evolving opportunities, and challenges with the speed, flexibility, and rigor required to address them. Discovery at such an accelerated pace continues to deliver transformative scientific and technological advances across the breadth of fundamental and applied science programs of the DOE mission, while seeding new scientific directions within them. As noted in the 2015 DOE Basic Energy Sciences Advisory Committee report on *Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science,* "success requires a sustained campaign of strategic investments that initiates new research thrusts, attracts and sustains a leading scientific workforce having the necessary skills, and provides the innovative instruments and tools with which to carry out the work." LDRD plays a major role in this campaign by allowing prompt investment directed at DOE mission challenges, while leveraging the world-class technical and scientific expertise of our national laboratories.

A sample of recent LDRD accomplishments is highlighted in this section, demonstrating discoveries in applied materials sciences, biosciences, and cosmology, as well as important developments in data analysis and materials characterization techniques. A 3D-printing platform for catalytic systems developed by LDRD has demonstrated a transformative method applicable to future clean-energy technologies. Furthermore, analytical and experimental methods supported by LDRD are providing key insights for the discovery, optimization, and processing of functional materials through real-time observation of their structures in a working environment, thus paving the path for innovations in energy production and storage, as well as in nuclear energy. Such methods are also unraveling the mysteries of structure–function relationships in bio-membranes relevant for membrane-targeting antibiotics and other therapeutics. Finally, computational tools developed by LDRD are enhancing our understanding from nanoscale form and structure in clean-energy materials to cosmological-scale processes, while establishing new paradigms for scientific innovation and discovery in general.

SCIENTIFIC INNOVATION & DISCOVERY

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52 Accelerated Materials Design Through First-Principles Calculations and Data Mining

54 Towards the Exascale Sky

Modulation Enhanced Diffraction: A New Tool for Powder Diffraction and Total Scattering Studies

Brookhaven National Laboratory

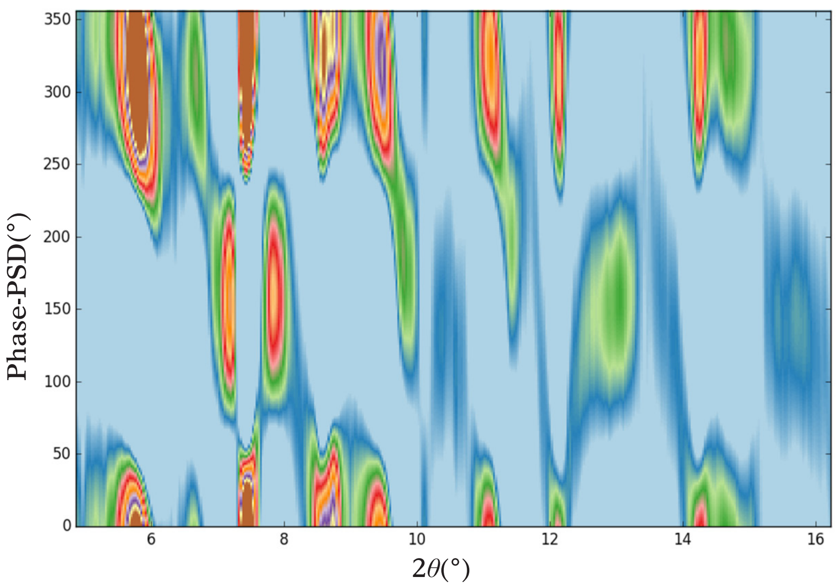
Tracking the structures of materials at work at the atomic scale

Project Overview

In addition to understanding static or steady-state atomic structures of materials, it is important to understand how they evolve during operation under real working conditions, such as an electrode material in a charging and discharging battery or a catalyst material functioning during the abatement of exhaust gases in vehicles. The Modulation Enhanced Diffraction (MED) method is being implemented at the National Synchrotron Light Source II as a new characterization tool to recover information buried in the larger signal emanating from the predominantly passive part of the structure.

Why It Matters

The National Synchrotron Light Source II is a new state-of-the-art synchrotron light source at Brookhaven National Laboratory that allows scientists to probe the fundamental properties of matter, paving the way to new scientific discoveries and innovations. This light source enables Brookhaven to explore transformational discovery through synchrotron science and operando and in situ energy science leadership. To determine the structural features responsible for key properties in a functional material, investigators need to track the time evolution of the material's structure in its working environment (operando) at the atomic scale. The aim of this project was to expand the in situ and operando time-dependent diffraction capabilities at the X-ray Powder Diffraction beam line and other diffraction and scattering beam lines at the National Synchrotron Light Source II.



Oxidation-reduction of a copper and cerium oxides catalyst in pulsing H_2/O_2 gases at 250°C in a constant stream of CO/O_2 mixture. The demodulated powder patterns as a function of phase angle is shown, where various copper and copper oxide phases can be distinctly identified.

MED is a well-established technique in spectroscopy, but very new in powder diffraction. This project, which developed procedures and algorithms to implement MED, stems from a collaboration between two national laboratories (Brookhaven and Argonne) and Stony Brook University using the National Synchrotron Light Source at Brookhaven and the Advanced Photon Source at Argonne. Structural selectivity is enhanced in MED by cycling an external parameter, such as the reactant composition, temperature, pressure, or electrical field during the course of the diffraction measurement.

For example, the relative proportions of copper (Cu) and copper oxide (Cu_xO) supported on ceria (CeO_2) are an important parameter in several catalysts. Procedures to implement MED were developed and benchmarked by studying the oxidation and reduction of a mixture of Cu and Cu_xO. The measurements were conducted by periodically pulsing hydrogen and carbon monoxide (H_2/CO) gases over the catalyst. When the catalyst goes from inert to active, MED can detect the small structural changes that allow for the catalyst to selectively remove CO from a mixture of CO and H_2, prior to input to fuel cells.

MED can uncover subtle changes normally undetected by conventional diffraction methods. Importantly, MED can be executed over the timescale of interest for most functional materials, which ranges from seconds to minutes or even hours.

Technical & Scientific Achievements

This project furthers the development of transformational materials for energy production and energy storage and for mitigating the environmental consequences of energy usage. Eric Dooryhee, the principal investigator on this project, says that "MED will become key to discovering, optimizing the synthesis of, and processing of transformational energy materials at the National Synchrotron Light Source II."

This project was crucial for benchmarking a new method (MED) that is likely to be applicable to industry-relevant processes, such as adsorption and desorption in membranes and filters, reversible ion intercalation in host matrices for batteries and fuel cells, polarization- or magnetization-induced structural shifts in piezoelectric actuators, and change of surface states in automotive catalysts under changing pressures of the reactants as well as for materials used in the nuclear industry.

Contributions & Impact

The software developed in this project is available at http://sourceforge.net/projects/twodmed/ and comprises two packages: 2DFLT (to edit the in situ data set) and 2DMED (to perform demodulation of the diffraction data). It is freeware and open source.

Publications

Caliandro, R., et al., "Static and Dynamical Structural Investigations of Metal-Oxide Nanocrystals by Powder X-Ray Diffraction: Colloidal Tungsten Oxide as a Case Study." *ChemPhysChem* **17**(5), 699–709 (2016).

Caliandro, R., et al., "Tailored Multivariate Analysis for Modulated Enhanced Diffraction." *J. Appl. Crystallogr.* **48**(6), 1679–1691 (2015).

Acknowledgments

This work was led by Brookhaven National Laboratory researchers Eric Dooryhee, Sanjit Ghose, Jose Rodriguez, Sanjaya Senanayake, Goknur Tutuncu, and Jonathan Hanson. Other collaborators included John Parise of Brookhaven and Stony Brook University, Andrey Yakovenko of Argonne National Laboratory, and Rocco Caliandro of Istituto di Cristallografia of Bari, Consiglio Nazionale delle Ricerche in Italy.

"MED will become key to discovering, optimizing the synthesis of, and processing of transformational energy materials."

Determining the Nanoscale Lateral Organization of Bio-Membranes Using Cold Neutrons

Oak Ridge National Laboratory

Researchers taking steps to discover how structure leads to biological function

Project Overview

Scientists at Oak Ridge National Laboratory are developing tools to show how membrane structure controls biological function, and to determine the in-plane organization of biologically relevant model membranes, even that of live bacteria. The experiments carried out at Oak Ridge's Spallation Neutron Source and High Flux Isotope Reactor rely on neutrons' unique ability to differentiate between atoms of hydrogen and deuterium, a heavy isotope of hydrogen.

Why It Matters

Cell membranes are composed mostly of lipids and proteins. The lipids form a fluid bilayer in which the proteins reside. Within the lipid bilayer, certain lipids associate with each other to form domains (see figure) that play an important role in the life of a cell. These functional domains—commonly known as lipid rafts—are thought to participate in a range of biological processes including the sorting of proteins, transport across the membrane, and entry and exit of viruses from cells. Despite their central role in biology, we have yet to observe lipid rafts in the membrane of a living organism.

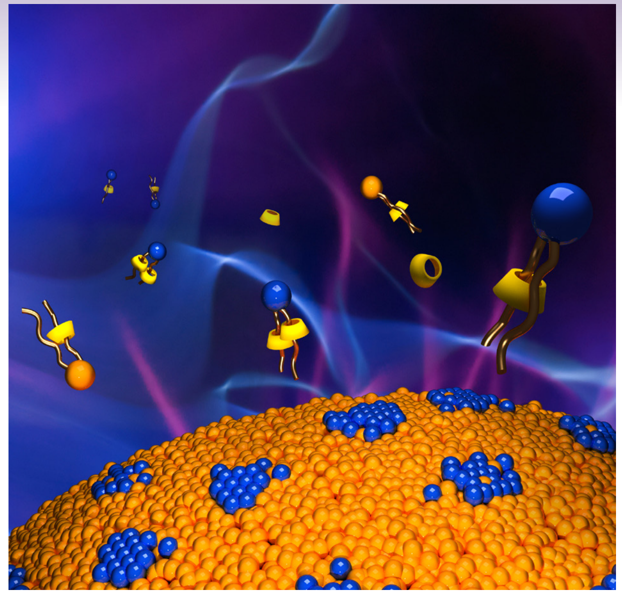
Although researchers suggested the concept of lipid domains in the early 1980s, they have eluded detection for almost 40 years. Despite the lack of direct experimental evidence, the emerging consensus is that the formation of functional lipid domains in cells occurs at the nanoscale. It is therefore not surprising that such small, and possibly ephemeral, structures have proven difficult to detect with traditional imaging techniques. Neutrons, however, are intrinsically nanoscopic probes with wavelengths that parallel the dimensions of the proposed lipid domains, offering a real possibility of detection.

Technical & Scientific Achievements

Oak Ridge developed the analytical and experimental tools to detect lipid domains in model membranes using intense cold neutron beams. Combining cold neutrons with isotopic labeling (namely hydrogen and deuterium), researchers unambiguously detected nanoscopic lipid domains in model membrane systems. Importantly, these domains seem to grow in size when biologically abundant lipids are substituted with ones not found in nature. Are lipid rafts truly nanometer-sized? In her *Journal of the American Chemical Society* "Spotlight" comment, the Seattle-based science writer and Fulbright fellow Deirdre Lockwood wrote, "By providing the best characterization yet of these tiny rafts, this study advances our understanding of how the composition of lipid bilayers regulates their physical organization and ultimately aids the critical process of signal transduction across a cell membrane."

Contributions & Impact

Having developed the analytical and experimental tools to detect nanoscopic lipid domains in model membranes, Oak Ridge researchers next embarked on detecting these features in a living bacterium, *Bacillus subtilis*. They generated specific neutron contrast in genetically manipulated cells (e.g., by incorporating hydrogen-labeled lipids into deuterated cells) grown under differing contrast (i.e., hydrogen and deuterium) conditions. Eventually, researchers will use small-angle neutron scattering to assess the presence or absence of nanoscopic and mesoscopic domains in live bacteria. The detection of lipid rafts in live cells will be a landmark in contemporary biology, and the development of the tool sets to unravel this longstanding question in biology will spur innovation of membrane-targeting antibiotics and therapeutics.



Cell membranes, as illustrated here, possess a complex 3D architecture, including nonrandom lipid lateral organization within the plane of a bilayer leaflet and compositional asymmetry between the two leaflets.

"The detection of lipid rafts in live cells will be a landmark in contemporary biology."

Publications

Heberle, F. A., et al., "Bilayer Thickness Mismatch Controls Domain Size in Model Membranes." *J. Am. Chem. Soc.* **135**(18), 6853–6859 (2013).

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Acknowledgments

Oak Ridge National Laboratory researcher J. Katsaras led this work, with significant contributions from F. A. Heberle, J. D. Nickels, R. F. Standaert, X. Cheng, J. G. Elkins, C. B. Stanley, and D. A. A. Myles. External collaborators included J. Pan (University of South Florida), G. W. Feigenson (Cornell University), and G. Pabst (University of Graz).

The Mechanisms of Soil Microbiome Response to Drought in Tropical Forests

Lawrence Berkeley National Laboratory

How drought induces metabolic changes in tropical soil microbes, altering carbon cycling

Project Overview

Microbes are key regulators of carbon storage in soils and provide critical nutrients for plant growth. Disturbances such as drought can change soil microbial metabolism and significantly impact soil carbon processing and plant productivity. Drought in humid tropical forests is a particular concern because of their importance to the global carbon cycle. In this project, researchers combined expertise in microbiology, systems biology, geochemistry, and ecosystem ecology to discover the mechanisms tropical soil microbes use to respond to repeated drought and to determine the consequences for soil carbon transformation.

Why It Matters

This research at Lawrence Berkeley National Laboratory demonstrated that even relatively small changes in soil water potential from simulated drought in tropical forests resulted in a large change in microbial diversity, community composition, metabolism, and carbon processing. “Significantly, despite a lack of previous drought in these wet tropical forests, native soil microbes were able to adapt and were more resistant to future episodes of drought,” explained Nicholas Bouskill, co-investigator for the project. However, these adaptations may come at a cost. The fraction of aromatic carbon in soil that is typically more resistant to microbial degradation decreased under drought conditions. This is important because accurate predictions of the future strength of Earth’s terrestrial carbon sink, which removes approximately a quarter of human-related carbon dioxide emissions, require that such microbial response mechanisms are represented in next-generation ecosystem models. However, “the vast numbers and diversity of soil microbes, the physical and chemical complexity of soils, and the need to study soil microbes as communities in their natural habitat, means that multidisciplinary teams are essential to decode their functioning,” said Eoin Brodie, principal investigator. Such multi-disciplinary approaches to understanding Earth’s microbiomes and their role in environmental stewardship and energy security has been heralded by the U.S. Office of Science and Technology under the recently announced National Microbiome Initiative. Building on this project and others, Lawrence Berkeley launched its ambitious Microbes-to-Biomes initiative to reveal, decode, and harness Earth’s critical microbiomes. This initiative will interface with other key DOE efforts to advance Earth-system modeling capabilities.

Technical & Scientific Achievements

Lawrence Berkeley developed new metabolomics approaches that provided the first glimpse into the metabolic response of soil microbes under drought stress in tropical forests, showing that soil microbes produced an array of chemicals (osmolytes) to protect themselves from the effects of drought. These chemicals require carbon and other nutrients, and the soil microbes produced extracellular enzymes to release those resources, in addition to producing other chemicals such as antibiotics to compete with other microbes. “These new metabolomic capabilities are uncovering the hidden world of microbial metabolism and communication in natural environments and are now being used to study how microbes make soil organic matter, and improve plant growth and the health of humans and animals,” said co-investigator Trent Northen.

Contributions & Impact

In many ecosystems around the world, climate change is creating new conditions that have not been previously experienced and it is unclear whether indigenous organisms have the capacity to adapt. Drought in typically wet tropical forests is a prime example because native vegetation can lack drought-tolerance traits, leading to large-scale tree mortality, species loss, and predictions that such forests may shift from carbon sinks to carbon sources. Before this project, nothing was known about the metabolic response of tropical soil microbes to drought and their ability to adapt. We now know that tropical soil microbes are very susceptible to drought, but can adapt and become more resistant to future incidents of drought. We know that cellular chemical protection is a key short-term microbial response to drought, but importantly, new communities of microbes emerge, producing antibiotics under drought stress and processing soil carbon in different ways that may impact soil carbon storage in these important carbon sinks.

Publications

Bouskill, N. J., et al., “Belowground Response to Drought in a Tropical Forest Soil. I. Changes in Microbial Functional Potential and Metabolism.” *Front. Microbiol.* **7**(333), 525 (2016).

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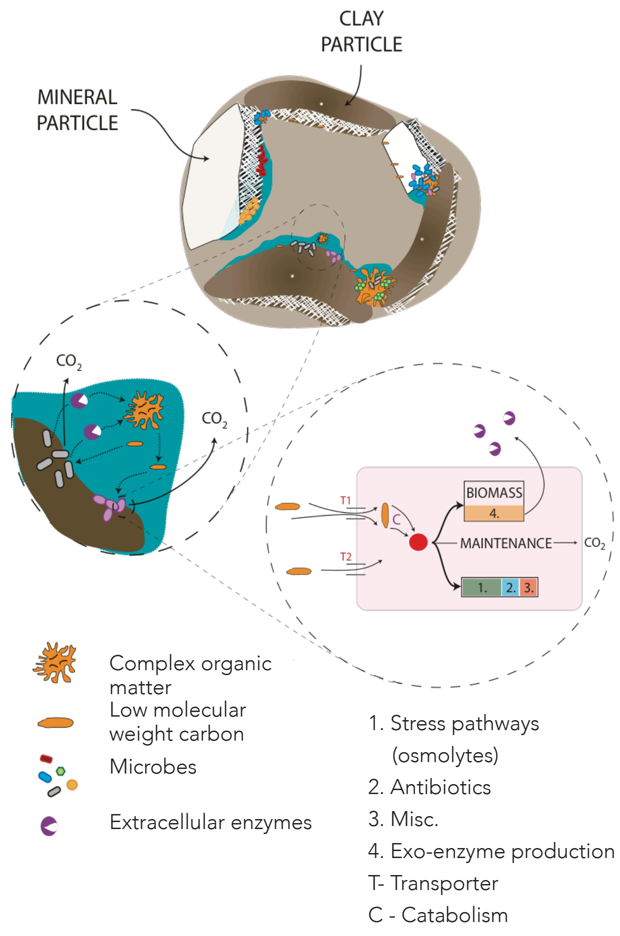
Acknowledgments

This work was lead by Lawrence Berkeley researchers Eoin Brodie, Nicholas Bouskill, Trent Northen, Zhao Hao, Peter Nico, and Benjamin Gilbert. External collaborators included Whendee Silver of the University of California, Berkeley; Tana Wood of the International Institute of Tropical Forestry; and Jizhong Zhou of the University of Oklahoma, Norman.



Reduced pore water volumes in tropical soils because of drought leads to higher solute concentrations and decreased resource diffusion to microbial cells. Microbes produce osmolytes, antibiotics, and extracellular enzymes in response.

“Native soil microbes were able to adapt and were more resistant to future episodes of drought.”

X Rays Reveal How High-Capacity Batteries Operate, and Fail

SLAC National Accelerator Laboratory

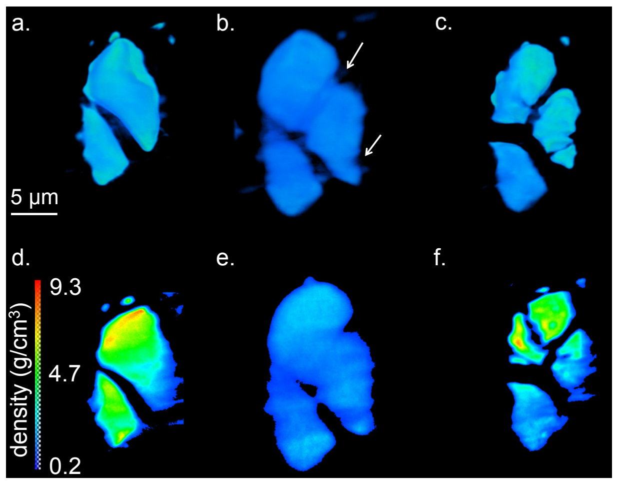
Real-time imaging showing electrode particles changing as lithium-ion batteries cycle

Project Overview

The shift from fossil fuels toward clean, renewable energy will require significant improvements in rechargeable battery technology for electric vehicles. The only way to achieve such dramatic improvements is to develop new materials for both electrodes and electrolytes. However, novel battery materials operate and fail in ways that are distinctly different from those now in use. These failure modes need to be carefully understood. This project harnessed high-energy x rays to directly observe and image novel electrode materials in lithium-ion batteries as they operated in real-world conditions.

Why It Matters

Lithium-ion battery technology has improved only slowly over time. Research has tended to focus on electrochemical measurements, which give only a general picture of a battery's health, or on ex situ measurements of individual components after a cell is disassembled. Researchers at the SLAC National Accelerator Laboratory used x rays to observe and measure batteries under realistic cycling conditions, allowing them to identify specific structural changes that lead to battery failure. With this information in hand, we can devise ways to avoid battery failure, and also conduct more systematic searches for electrode materials that continue to perform at high capacity over the lifetime of an electric vehicle.



3D images of germanium particles (a) before cycling, (b) after lithium-ion insertion, and (c) after lithium-ion removal. Cracks formed during cycling (arrows) completely fractured the largest particle.

Technical & Scientific Achievements

Researchers from the Stanford Synchrotron Radiation Lightsource, in collaboration with Stanford University, used transmission x-ray microscopy and x-ray diffraction to observe how high-capacity battery materials—including silicon- and germanium-based anodes and sulfur-based cathodes—fail during lithium-ion battery operation. High-energy x rays are powerful enough to penetrate many materials, allowing researchers to observe typical failure mechanisms in standard battery cells with few modifications.

High-capacity anode materials take up and release a high concentration of lithium ions during battery cycling. But the resulting swelling and shrinking can crack the electrode material and degrade its performance.

In this study, tomography was used to compile 3D images of germanium anode particles during charging and discharging, revealing the first unambiguous evidence of an anode material fracturing into completely isolated pieces. The 3D information also allowed researchers to calculate changes in particle density and volume. The density changes correlated to an incomplete flow of lithium ions out of the anode and a partial transformation of the initial germanium into a $Li_{15}Ge_4$-like phase during lithium-ion insertion. These observations can be linked to capacity loss during the first cycle of the battery, thus identifying the fracturing and incomplete delithiation as significant culprits.

Researchers also observed that the cycling behavior of individual germanium particles depended significantly on their size. During the first cycle, only particles with diameters larger than a few microns showed cracks and smaller particles expanded before their larger counterparts. However, by the second cycle, most particles lost electrical contact and no longer participated in charging—only the very largest germanium particles continued to expand and shrink. The deactivation of most of the particles after the first cycle accounts for at least a quarter of the total capacity loss observed in the first cycle.

Contributions & Impact

This work by Stanford Synchrotron Radiation Lightsource researchers at SLAC has developed a methodology for understanding the operation of complex, heterogeneous functional materials, which will enable advanced approaches to material synthesis. This methodology is one of the enabling technologies for transformative advances in imaging capabilities across multiple scales to solve longstanding challenges in the relationship between the structure of inhomogeneous matter and its behavior.

Publications

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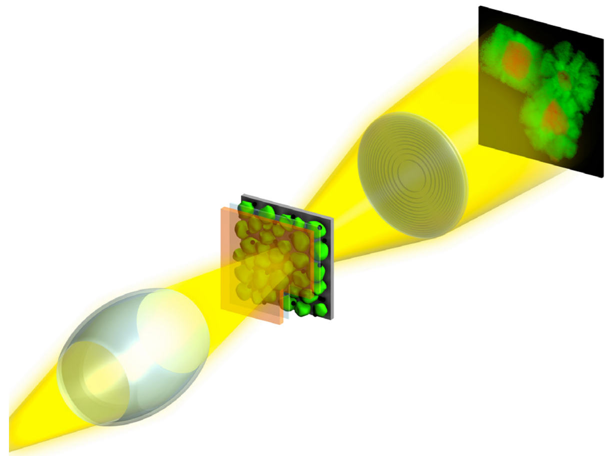
Acknowledgments

This work was led by Johanna Nelson Weker and Michael F. Toney at SLAC National Accelerator Laboratory with key contributions from Nian Liu and Yi Cui at Stanford University.



With transmission x-ray microscopy, researchers can image individual electrode particles and observe chemical and structural changes as a battery operates under realistic conditions.

"With this information in hand, we can devise ways to avoid battery failure."

Customized Assembly of Catalytic Systems by 3D Printing Technology

Ames National Laboratory

Using 3D printing of both reactor structures and catalysts themselves to study several chemical reactions

Project Overview

This initiative aims to integrate the assembly of nanostructured materials into mesoscopic catalytic arrays with the manufacture of macroscale chemical reactor vessels. Researchers at Ames National Laboratory are developing functionalized catalyst nanoparticle inks and printing them into 3D-printed reactor structures using additive manufacturing technologies. This will enable simple and efficient customization of catalytic setups in the laboratory, and facilitate the translation of fundamental catalyst discovery into process chemistry for industrially relevant conversions. The work aligns with Ames' strategic initiative of greener advances for catalysis for energy.

Why It Matters

The DOE's Basic Energy Sciences Advisory Committee identified a set of opportunities in mesoscale sciences for clean energy technologies. One of the priority directions is directing the assembly of hierarchical functional materials, along with their grand challenge for catalysis for energy: the design and controlled synthesis of catalytic structures. These issues can be addressed by the merger of novel top-down additive manufacturing technologies with the bottom-up mesoscale self-assembly of nanostructured catalytic materials, which is the central goal of this project.

Technical & Scientific Achievements

Combining top-down 3D printing with bottom-up nanometer-scale structuring techniques was successfully achieved to produce reactors and active catalysts. Ames produced a pressure reactor by computer-aided design, shown in figure (a), that proved capable of working under 5-bar hydrogen gas pressure. Several iterations were performed on the reactor design and printing, with significant improvements at each step, proving the relevance of computer-aided design and on-site production. The final reactor includes gaskets, ports for pressure gauges, liner, pressure relief valves, and a catalyst bed with defined geometry incorporated in the path of the reactive gas. A novel concept of replaceable catalyst cap inserts was conceived and developed into working systems to highlight advantages of computer-aided catalyst production, shown in figure (b). Catalysts for five different reactions were 3D printed and three proved to be active. Researchers demonstrated incorporation of nanostructured catalysts into 3D-printed material via scanning electron microscopy and catalytic activity tests.

Contributions & Impact

The project team produced a versatile reactor and catalyst platform—in particular, three catalytic reactions were demonstrated using 3D-printed cataysts. The platform and demonstration should have an impact in future scientific research and industrial development where simple laboratory setups will allow for efficient study of catalyst behavior.

Publications

Fleckenstein, J. E., "Supported Metal Complexes: From Mesoporous Silica to 3D Printed Polymers." Iowa State University M.Sc. Thesis, Ames, Iowa (2015).

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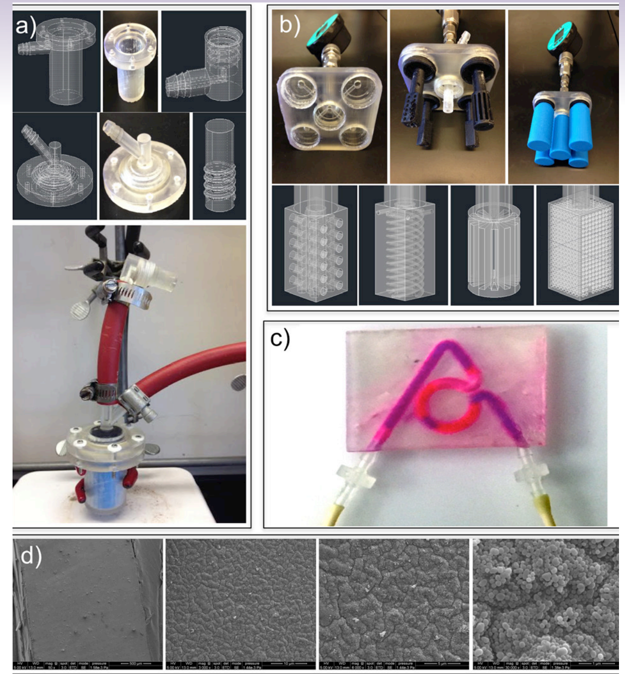
Acknowledgments

This project was supported by the Ames Laboratory's LDRD program through the Division of Chemical and Biological Sciences. The work at Ames was led by Igor I. Slowing and Aaron D. Sadow, with key contributions from Jacob Fleckenstein, Sebastián Manzano, and Zachary Weinstein.



(a) 3D-printed pressure reactor containing a 3D-printed catalyst. (b) 3D-printed, four-port catalytic reactor for testing catalyst bed configurations, with computer-aided designs of catalyst cap inserts. (c) Catalytic example reactions performed with printed catalyst products. (d) Scanning electron microscopy images at increasing magnification showing the incorporation of a nanostructured mesoporous silica nanoparticle catalyst in a 3D-printed material.

"This will enable simple and efficient customization of catalytic setups in the laboratory, and facilitate the translation of fundamental catalyst discovery into process chemistry for industrially relevant conversions."

Accelerated Materials Design Through First-Principles Calculations and Data Mining

Lawrence Berkeley National Laboratory

Quantum chemistry calculations performed using state-of-the-art supercomputers to drive the discovery of new materials

Project Overview

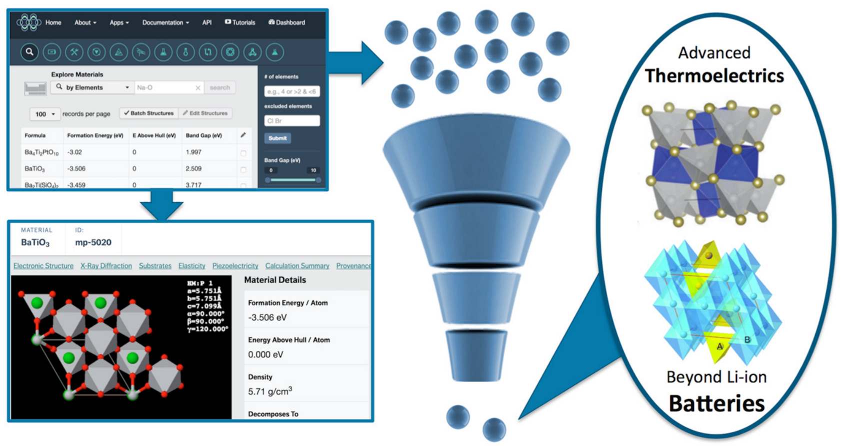
The Materials Project at Lawrence Berkeley National Laboratory aims to accelerate the discovery of new materials for applications ranging from next-generation batteries to advanced solar panels by providing the materials science community with extensive data from state-of-the-art simulations conducted at top supercomputing centers.

Why It Matters

New materials are key to both improving the efficiency of our existing energy infrastructure and enabling the design of paradigm-changing technologies that will help ensure the security and sustainability of our energy resources. With recent advancements in supercomputing hardware, it is now possible to calculate the properties of nearly every known inorganic material using algorithms for quantum chemistry like those from density functional theory. The Materials Project uses these tools to generate data and screen materials to find those that should be tested in efforts to make higher-capacity batteries, longer-lasting solar cells, more efficient thermoelectrics, and more active catalysts. Furthermore, data from a high-throughput approach can be used to learn how the structure of atoms and electrons in crystals relate to their measured properties, enabling researchers to design materials atom by atom, before ever running an experiment.

Technical & Scientific Achievements

The Materials Project has calculated the phase stability of over 65,000 inorganic compounds, along with 40,000 band structures, 3,000 full-elastic tensors, and nearly 1,000 piezoelectric tensors.



A "Google" for materials. Data for over 60,000 materials are stored at materialsproject.org, which can be filtered to determine the most suitable candidates for a given technology.

It has developed an advanced interface that users can access via both the Web and an application-programming interface. So far, the data and approach have been used to discover novel thermoelectric materials, reveal unique photocatalytic activity in known materials that may eventually be used to convert solar energy into chemical fuels, and to elucidate formation pathways that provide insights on how different materials might be synthesized.

Contributions & Impact

The Materials Project is contributing to a rapidly growing new paradigm in materials science in which computer simulations can inspire new materials and provide valuable insights into fundamental scientific questions about how the arrangement of atoms and electrons in a crystal relate to their behavior in modern devices. The online interface to The Materials Project has over 20,000 users from around the world, and each of the code bases used to perform the high-throughput calculations stored in the database are open source and actively developed by a team spanning numerous research groups. Using the Web interface, users can explore x-ray diffraction data, nanoparticle morphologies, nanoporous adsorbents, and molecular reduction–oxidation flow-battery chemistries, in addition to basic thermodynamic properties from density-functional theory simulations. Most recently, this infrastructure has even been adapted to accept user-submitted crystal structures to be simulated and stored in the public database.

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Acknowledgments

This work was intellectually led by the Materials Project team, and initial funding was provided by the LDRD program of Lawrence Berkeley National Laboratory. The contributions of numerous external collaborators and projects are gratefully acknowledged.

"A high-throughput approach can be used to learn how the structure of atoms and electrons in crystals relate to their measured properties, enabling researchers to design new materials atom by atom, before ever running an experiment."

Towards the Exascale Sky

Argonne National Laboratory

Extreme-scale computing enabling high-resolution simulations of the universe

Project Overview

Next-generation observations in astronomy and astrophysics promise an unprecedented jump in our knowledge of the universe. Extracting new science from those observations requires a theoretical and computational understanding and a modeling capability that is revolutionary. Work at Argonne National Laboratory will lead to the development of the first exascale computational cosmology capability for next-generation sky surveys. Exascale computation implies at least a quintillion floating-point operations per second—a thousand petaflops. Modern supercomputers are on the developmental pathway to the exascale. Researchers are creating a capability to exploit next-generation computational architectures that are aligned with the scientific needs of fields such as cosmology and accelerator and plasma physics.

Why It Matters

This research is directly relevant to the goals of DOE's Cosmic Frontier program in the Office of High Energy Physics. According to Salman Habib, project leader and a senior member of the Kavli Institute for Cosmological Physics at the University of Chicago, "the unveiling of the large-scale universe by sky surveys has transformed cosmology, fundamental physics, and astrophysics." Dark energy (the proposed cause of the accelerating expansion of the universe) and dark matter (the unknown form of matter required to explain observations related to the role of gravity in the universe) are at the center of an intense global effort to fathom the mysteries posed by their existence. Large-scale simulations play a central role in this enterprise. They are a source of computational discoveries of new effects, design of new cosmological probes, and high-accuracy predictions for many cosmological models. Large-scale simulations also provide high-fidelity synthetic catalogs for testing, validating, and optimizing observational strategies. Habib explains that "the high-performance computing and data-intensive computing advances that underlie this project will also have broad impact in several fields beyond cosmology, including accelerator and plasma physics, computational co-design, particle-transport simulations, and management and analysis of very large data sets."

Technical & Scientific Achievements

This project developed a leading-edge computational capability for large-scale cosmological simulations and their analysis. The HACC software (Hardware/Hybrid Accelerated Cosmology Code) was fully implemented on Mira (Argonne's 10-petaflop IBM Blue Gene/Q system). It broke the 10-petaflop performance barrier for the first time on the Sequoia system at Lawrence Livermore National Laboratory. An in situ analysis framework for HACC was developed that allowed the code to run analysis tasks at the same time as the main simulation. The code was then ported to Titan, the heterogeneous supercomputer at Oak Ridge National Laboratory, with the code kernel performance exceeding 20 petaflops. A new task-based load balancer was developed for this version of HACC. This scheme was used very successfully at the Oak Ridge Leadership Computing Facility and resulted in one of the world's largest high-resolution cosmological *N*-body simulations. New results on the effects of neutrino mass on galaxy cluster abundance were obtained following this work on implementation of neutrinos in HACC. It was shown that the cluster abundance depends on the neutrino mass following a universal form of the mass distribution function. This result makes inferences from observations of clusters (especially observations of high redshift clusters with the South Pole Telescope) much more straightforward. A technique for handling this inference problem followed from earlier work in this project on the cluster and galactic halo concentration–mass relationship.

Contributions & Impact

The improved HACC that resulted from this work has been used for a number of additional scientific studies including precision emulation of cosmological observables, creation of synthetic galaxy catalogs for cosmological surveys, and for investigations of the Sunyaev-Zel'dovich effect (the distortion of the cosmic microwave background spectrum from background photons scattering from energetic electrons in and around galaxy clusters). The HACC simulations played an important role in the measurement of the pairwise kinematic Sunyaev-Zel'dovich effect (i.e., the influence on the Sunyaev-Zel'dovich spectrum from motion of the clusters with respect to the cosmic microwave background rest frame) by the joint efforts of the South Pole Telescope at the National Science Foundation South Pole research station and the Dark Energy Survey in the Chilean Andes. This project's work on large-scale data analytics was continued with support from Lawrence Berkeley National Laboratory's National Energy Research Scientific Computing Center, where investigators have access to large storage and analysis capabilities for post-simulation data processing.

Publications

Heitmann, K., E., et al., "The Coyote Universe Extended: Precision Emulation of the Matter Power Spectrum." *Astrophys. J.* **780**(1), 111 (2014).

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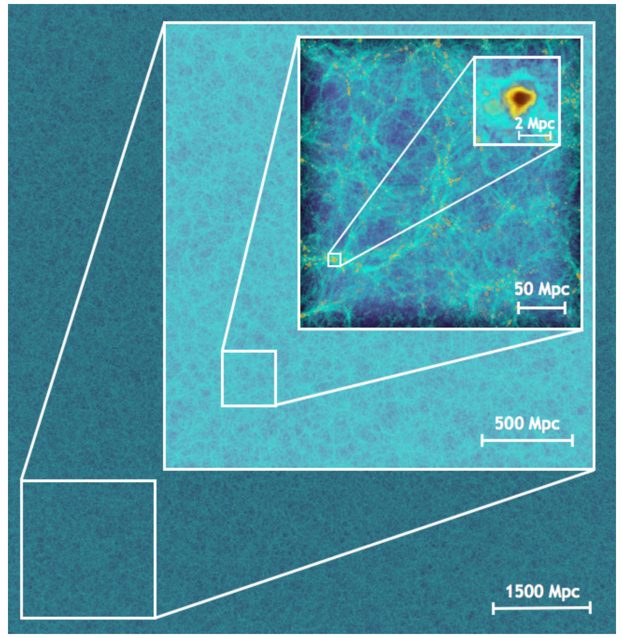
Acknowledgments

External collaborators included Martin White (University of Caifornia, Berkeley) who made synthetic catalogs from HACC simulations for the Dark Energy Spectroscopic Instrument, and Nikhil Padmanabhan (Yale University) who collaborated on the MockBOSS suite of simulations for the Baryon Oscillation Spectroscopic Survey. We acknowledge the contributions of Vitali Morozov (Argonne Leadership Computing Facility) for critical work on performance enhancement for the HACC computational kernels.



Multi-resolution view of the trillion-particle "Outer Rim" simulation using HACC (Hardware/Hybrid Accelerated Cosmology Code). One of the world's largest cosmological simulations, the (linear) box size is 4,225 megaparsecs (about 14 billion light-years).

"This project developed a leading-edge computational capability for large-scale cosmological simulations and their analysis."

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