Lawrence Livermore National Laboratory

January/February 2022

Science & Technology REVIEW

GETTING TO CARBON NEUTRAL

Also in this issue:

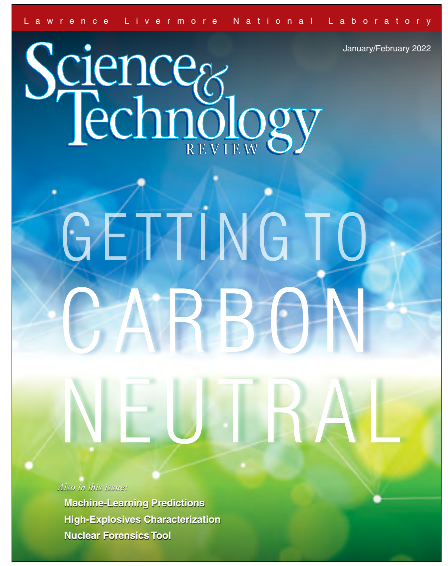
Machine-Learning Predictions

High-Explosives Characterization

Nuclear Forensics Tool

About the Cover

Achieving carbon neutrality requires a well-considered combination of technologies, private involvement, and public policies. As described in the article beginning on p. 4, Lawrence Livermore National Laboratory developed the *Getting to Neutral: Options for Negative Carbon Emissions in California* report, a three-pillared approach to bridge the gap between California's initial plans to reduce carbon emissions and the state's goal to achieve net zero emissions by 2045. The cover image represents the network of strategies and stakeholders involved in achieving carbon neutrality for the environment.



Cover design: Mark Gartland

About *S&TR*

At Lawrence Livermore National Laboratory, we focus on science and technology research to ensure our nation's security. We also apply that expertise to solve other important national problems in energy, bioscience, and the environment. *Science & Technology Review* is published eight times a year to communicate, to a broad audience, the Laboratory's scientific and technological accomplishments in fulfilling its primary missions. The publication's goal is to help readers understand these accomplishments and appreciate their value to the individual citizen, the nation, and the world.

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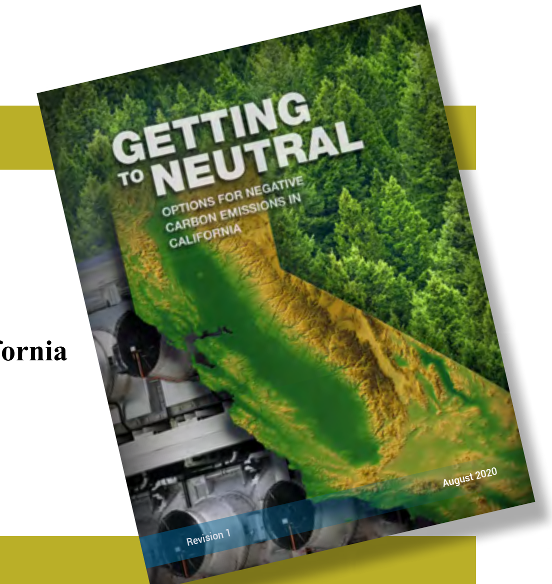
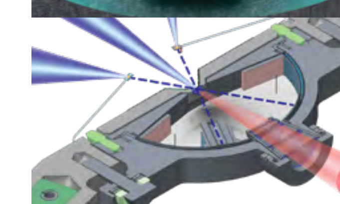
Contents



Feature

Research Highlights



Departments

Optimized Spectrometer Design

Scientists at Lawrence Livermore National Laboratory collaborated with Princeton Plasma Physics Laboratory to design a novel x-ray crystal spectrometer. The device provides high-resolution measurements of a challenging feature of high-energy-density (HED) matter produced by National Ignition Facility (NIF) experiments. The work is featured in the May 10, 2021, online edition of *Review of Scientific Instruments*.

A spectrometer built earlier for NIF measures profiles of key parameters such as ion and electron temperatures in large volumes of hot plasmas magnetically confined in doughnut-shaped tokamak fusion devices to facilitate fusion reactions. NIF's laser-produced HED plasmas, by contrast, are tiny, point-like substances. The newly developed multi-optics high-resolution absorption x-ray spectrometer (HiRAXS) uses crystals for extended x-ray absorption fine structure (EXAFS) experiments examining copper, tantalum, and lead. X-ray energy increases and signal noise decreases from copper to tantalum to lead, motivating efforts to optimize spectrometer design.

"Experiments at NIF that measure the EXAFS spectrum at high x-ray energies have had low signals," says Marilyn Schneider, Livermore's Radiative Properties group lead and a co-author of the paper. "The HiRAXS design concentrates the low signal and increases the signal-to-noise ratio while maintaining the high resolution required for observing EXAFS." Co-author Yuan Ping adds, "The new design enables us to meet strict requirements for EXAFS measurements to prove the thermal state of highly compressed, high-atomic-number materials."

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Quantum Computing Stability

Livermore physicist Jonathan DuBois and collaborators proved that quantum computing errors are tied to cosmic rays. Understanding the cause of errors and how quantum circuits react to them is required to build a functioning quantum system and move closer to achieving the potential of quantum computing.

The team's findings, published June 16, 2021, in *Nature*, indicated that fluctuations in the electrical charge of quantum bits (qubits) can be highly correlated rather than independent and random as assumed in earlier approaches to error correction. Bursts of energy from outside the system, such as cosmic rays, can affect nearby qubits by creating a blast of high-energy electrons, potentially heating the quantum device's substrate and disrupting the qubits. When particle impact occurs, electrons scatter and produce high-energy vibrations and heat, further altering the electrical field and the thermal and vibrational environment around the qubits. As a result, correlated errors span the system and simultaneously affect performance.

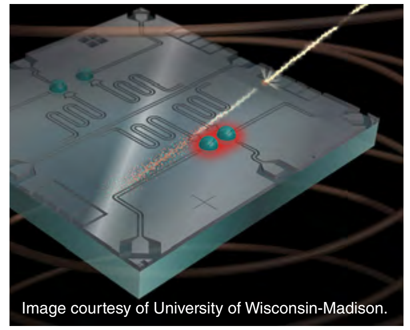
Image courtesy of University of Wisconsin-Madison.

To view the disruptions, researchers sent radio frequency signals into a four-qubit system. By measuring the excitation spectrum and performing spectroscopy, the team saw the qubits "flip" from one quantum state to another at the same time in response to changes in the charge environment. The team concluded that mitigation strategies must be developed to protect quantum systems from correlated errors due to cosmic rays and other particle impacts. Says DuBois, "We are focused on understanding the dynamics of these microscopic explosions and developing ways to absorb the energy before the delicate states inside quantum computing devices are disrupted." Research partners included the University of Wisconsin-Madison, Fermi National Accelerator Laboratory, Google, Stanford University, Kavli Institute for Cosmological Physics, Institute of Nuclear Physics Sezione di Roma, and Sorbonne Université.

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Evolution of Liquid and Solid Microjets

Lawrence Livermore National Laboratory scientists tested and confirmed predictions of a computational study investigating the effect of melting on shock-driven metal microjets. Livermore scientist David Bober sought to test a surprising trend reported in the 2020 simulations. "The computational study predicted that melting the base material might not always lead to a dramatic increase in the mass of material ejected from a surface feature, which goes against the conventional wisdom," he says. Bober and Livermore scientists Kyle Mackay, Minta Akin, and Fady Najjar co-authored a manuscript detailing their experimental results in the July 2021 *Journal of Applied Physics*.

In the experiment, the team applied a fast-moving projectile to a tin plate, initiating a fluid-like jet of tin into the path of an intense x-ray beam. Images using x rays and an array of high-speed cameras enabled the team to calculate the jet's mass and velocity, confirming the 2020 simulations. Colleagues at Argonne National Laboratory's Advanced Photon Source Dynamic Compression Sector contributed to the success of the experiment.

The study of microjets supports research in broader jetting and ejecta processes that occur throughout condensed matter shock physics, from explosives to asteroid impact. Future shots have been planned to explore the phenomena. "We are on a path to improving ejecta models by detailing the physics that happen around the melt transition," says Bober.

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A New Age for Public-Facing Science at Lawrence Livermore

READERS of this issue's feature article, beginning on p. 4, will find a phrase not often seen in conjunction with science at Lawrence Livermore National Laboratory: "With funding from the Livermore Lab Foundation through a generous donation from the ClimateWorks Foundation—philanthropic organizations supporting scientific research..." When I first heard that the Livermore Lab Foundation (LLF), an independent 501(c)(3) nonprofit, had been established, I thought it would never impact my research life at the Laboratory. Now, LLF, working through the Laboratory's Strategic Partnership Projects Program, has become a valuable partner helping us pursue important new science. The foundation's support of projects at the Laboratory gets important work out to the public—from providing funding mechanisms to developing communications tools and classroom materials to cultivating new public–private partnerships.

Science-oriented foundations such as ClimateWorks are eager to partner with Lawrence Livermore to investigate new problems that have not yet risen to full national attention. In the case of *Getting to Neutral*, ClimateWorks knew about the Laboratory's work on negative emissions technology and felt assured their gift to the LLF would encourage the kind of analysis they believed California needed. The *Getting to Neutral* report has been extremely influential in both the California and national plans for climate response. The scientists who wrote the report have shared their findings and insights with state and federal policymakers seeking a national carbon reduction. Companies developing the technologies that can accelerate the plan's success have been brought into the conversation with stakeholders. As noted in the article, Microsoft Corporation was inspired by the report's findings to develop a carbon-negative plan, and other companies have sought Livermore expertise to add carbon removal to their own plans to reduce carbon emissions. A major Department of Energy program to analyze the nation's approach to carbon neutrality has been launched, led by our Laboratory. A communication toolkit created by Lawrence Livermore and LLF will further grow the conversation in the community and with students. The seeds sown by this philanthropic effort are flourishing.

Today's research ecosystem is an expanded model of public–private partnerships and collaboration that includes the philanthropic sector. The LLF and other nonprofit organizations provide an agile mechanism to facilitate philanthropic gifts, promote the Laboratory in new communities, advance our science and technology goals and capabilities via new research opportunities, and amplify the societal impact of Livermore Laboratory's research and development.

The most likely place for philanthropy to look to our Laboratory is in public-facing science. Our deep bench of scientists and technologists can address many problems that philanthropies care about, from climate change to health and contagious disease. And because we care very much about those problems, our scientists are eager to contribute to the public discourse. *Science & Technology Review* complements philanthropic and public interest by presenting Lawrence Livermore science—from all funding sources—in the pages of every issue to underscore the value of all research underway at the Laboratory.

Three diverse highlights kick off 2022's review of Livermore science and technology. The article starting on p. 12 describes advances in artificial intelligence designed to ensure more accurate predictions in machine-learning applications applied to fields as diverse as national security and future smart car operations. A highlight beginning on p. 16 describes the union of Livermore core competencies and one-of-a-kind capabilities at the National Ignition Facility to expand materials research supporting the stockpile stewardship mission. The final highlight starting on p. 20 presents a prototype device developed to support public safety and national security by analyzing debris following a nuclear event.

■ Roger Aines leads the Director's Initiative in Engineering the Carbon Economy.

S&TR January/February 2022

THE PATH TO A CARBON NEUTRAL CALIFORNIA

A Livermore report outlines a strategy to reduce California's carbon emissions to net zero by 2045.

AN executive order signed in 2018 by then-Governor Jerry Brown set the ambitious goal to make California carbon neutral by 2045. To be carbon neutral, the state must achieve zero net emissions of climate change–causing gases, the most abundant of which is carbon dioxide (CO_2). The executive order set agencies such as the California Air Resources Board (CARB) in motion developing pathways to reduce the state's emissions by adopting low-carbon fuels and accelerating deployment of renewable energy, energy efficiency, and electrification of the state's buildings and vehicles, among other strategies. CARB's carbon-modeling reduction scenarios indicate that at least an 80 percent reduction can be achieved by 2045. But getting the state to true carbon neutrality would require additional measures and technologies.

At the same time, William Goldstein, Lawrence Livermore National Laboratory's director in 2018, established the "Engineering the Carbon Economy" initiative. This work is directed at developing technologies to reduce the world's atmospheric CO_2 by 25 percent. The reduction is required to constrain global warming to 2°C, even after achieving carbon-free electricity and emissions-free industry and transportation. (See *S&TR,* July/August 2019, pp. 4–11.) "We realized that huge amounts of carbon removal were needed to hit this target, making it difficult to achieve," recalls Roger Aines, leader of the Director's Initiative in Engineering the Carbon Economy. "We asked ourselves how we could combine different measures to achieve this reduction."

California policymakers asked the Laboratory to prepare a report on carbon removal solutions. With funding from the ClimateWorks Foundation and the Livermore Lab Foundation—philanthropic organizations supporting scientific research—a team of Laboratory and academic researchers released *Getting to Neutral: Options for Negative Carbon Emissions in California* in 2020. The report demonstrated how the state could achieve the additional 20 percent reduction identified in CARB's study and reach the 2045 carbon neutrality goal. Better yet, the goal could be met through "negative emissions," at modest cost, using resources and jobs within the state.

The Positives of Negative Emissions

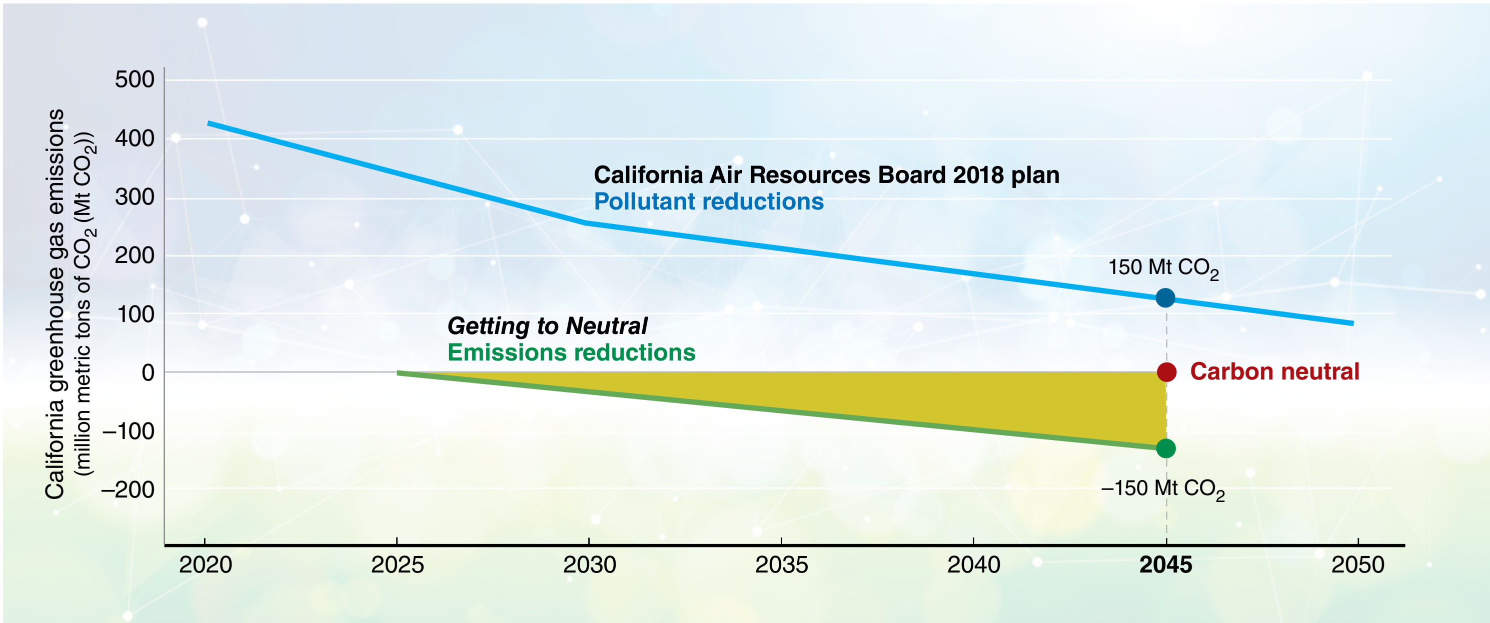
Materials scientist Sarah Baker, the first author of the report and leader of Livermore's Materials for Energy and Climate Security group, says, "We define negative emissions as CO_2 that is physically removed from the atmosphere through such approaches as biomass growth or direct air capture. Negative emissions do not include reductions in current or projected emissions and require additional effort." Drawing from a substantial body of previous studies in the scientific literature, *Getting to Neutral* reached the following conclusion: "By increasing the uptake of carbon in its natural and working lands, converting waste biomass into fuels, and removing CO_2 directly from the atmosphere with purpose-built machines, California can remove on the order of 125 million metric tons (Mt) of CO_2 per year from the atmosphere by 2045…" This strategy complements the state's current plan to reduce CO_2 emissions from 431 million metric tons of CO_2 (Mt CO_2) to 86 Mt CO_2 to achieve carbon neutrality by 2045.

The report added that negative emissions can be achieved with measures taken entirely within California by spending an estimated $10 billion per year, less than 0.4 percent of the state gross domestic product, on local jobs and industry. The strategy offers revenue opportunities for the private sector and makes out-of-state carbon offset purchases unnecessary. Negative emissions measures provide environmental co-benefits as well, helping to improve air and water quality, protect life and property, and increase soil health.

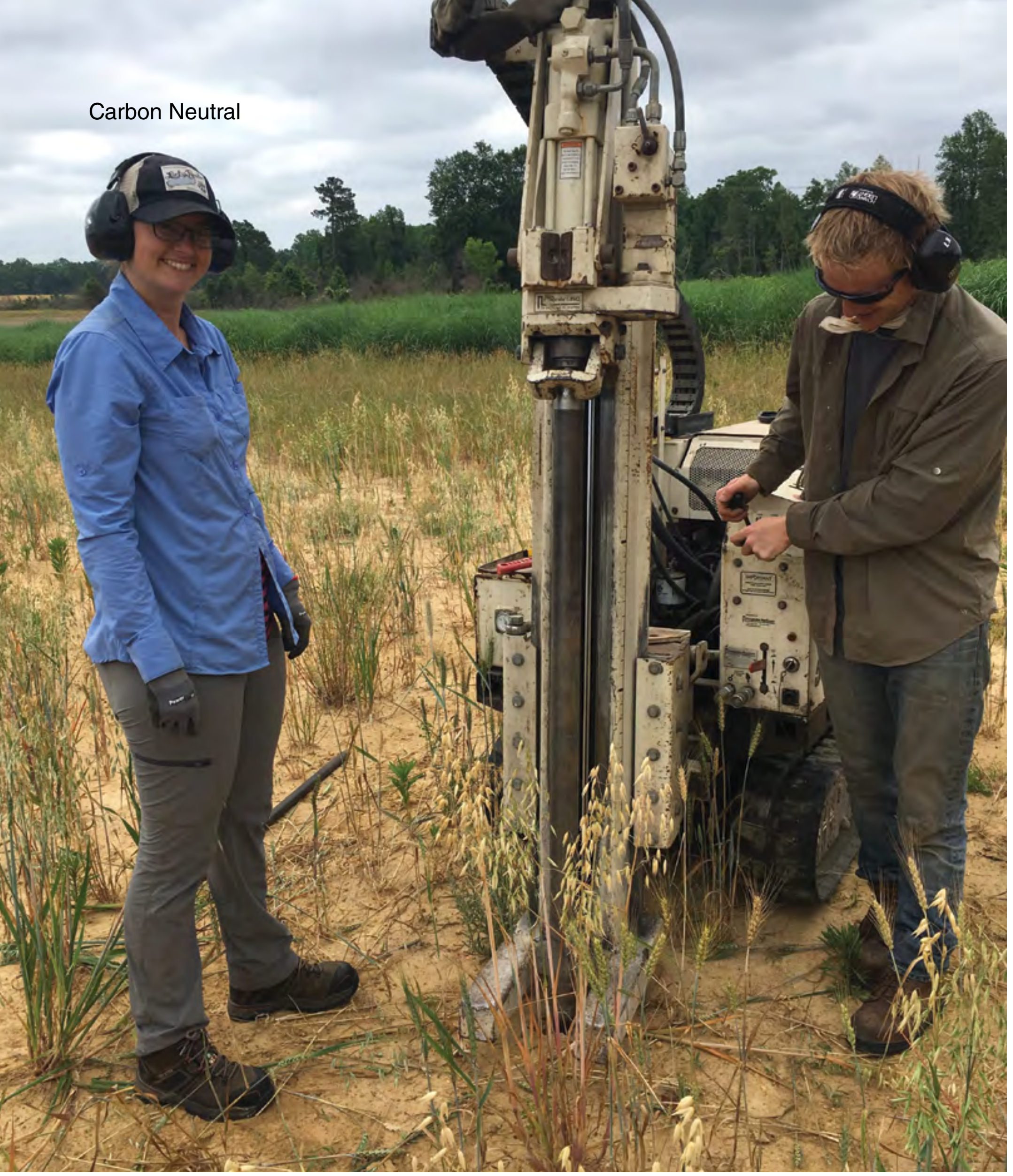
The report lays out a three-pillar approach to carbon reduction: removing CO_2 from the atmosphere via natural solutions (soils and forests); converting waste biomass to fuels and other products while storing the carbon underground; and capturing CO_2 from ambient air and then storing it underground.

Pillar 1: Natural Solutions

"Natural solutions remove CO_2 by storing carbon in plant biomass and soils," explains geochemist Jennifer Pett-Ridge, who leads the Environmental Isotope Systems group. Approaches include forest management practices, reforesting wildfire-disturbed areas, restoring grassland and tidal and freshwater wetlands, changing plant cover to reduce fallow periods on agricultural lands, and planting deep-rooted perennial plants. Forest management practices can increase carbon stocks above the amount currently stored, for example, by increasing harvest rotation length in managed forests, or actively replanting trees on deforested lands and in urban areas. In agricultural landscapes, restoring wetlands that can sequester carbon in peat soils and improve grassland management can also remove atmospheric CO_2. Worldwide, soils have lost an estimated 130 billion tons of organic carbon (equivalent to 480 billion tons of CO_2) to the atmosphere due to agriculture. Researchers project that soils could potentially store 1 billion tons of



The *Getting to Neutral* report proposes means to remove greenhouse gas emissions from the atmosphere that, when combined with the State of California's 2018 plan to reduce pollutants, would achieve carbon neutrality by 2045, unlike implementing the state plan alone.



Livermore's Erin Nuccio and Eric Slessarev use a Geoprobe coring device to collect soil cores from underneath shallow-rooted annual crops in North Carolina. Sample analysis helps the researchers determine how different plants—annual crops versus deep-rooted perennial grasses—effect soil carbon stocks.

carbon per year globally, while increasing soil fertility.

Pett-Ridge explains that no-till agriculture (not plowing fields), cover-cropping (never leaving the ground bare by planting alfalfa or other nitrogen-fixing crops), and planting perennial plants that keep roots in the ground year-round, all help to keep carbon in the soil and may increase the soil carbon amount. Planting perennials in agricultural lands also adds carbon because perennials' deep roots, which need less irrigation or fertilizer, distribute carbon throughout a large volume of the soil profile. "The deeper in the soil column the carbon is, the safer it is," says Pett-Ridge.

To estimate soil-carbon storage in California, postdoctoral researcher Eric Slessarev applied a set of simulated scenarios using the COMET planner, a database of biogeochemical model results that reports soil conservation practices and associated climate benefits on a per-county basis. Extrapolating for the state's soils, Slessarev scaled up COMET's scenarios to estimate the maximum technical potential for storing carbon in California soils, applying land use constraints such as the total amount of farmland in the state. The estimates represent a total possible technical benefit but do not consider other concerns, such as agricultural policies, the willingness of landowners to participate, and opportunity costs for using the land for other purposes.

The result: total emissions reductions from applying all conservation practices equaled 3.9 million tons of CO_2 equivalent per year (Mt CO_2e/yr). However, tracking how much carbon is stored and how long it will persist underground is difficult. "Large uncertainties are associated with the estimate," says Slessarev. "To improve the model, we need more data. As we add carbon to soils, we'll need to monitor how much goes in, what happens to the carbon, and whether the amount of greenhouse gases escaping the soil surface changes. We will learn as we go about what works and what doesn't." Slessarev, now a Livermore staff scientist, is leading a Laboratory Directed Research and Development Program–funded project to develop and test a global-scale model that simulates how soil chemical properties influence the capacity of soils to store carbon and improve the predictive power of climate change and Earth system models.

Natural solutions have advantages over more engineered approaches to CO_2 capture. "They require relatively lower investment and could start quickly—within a year or two," says Pett-Ridge. "These approaches to CO_2 removal are by far the cheapest, at least in California, based on per ton cost." Excluding economic and policy factors such as compliance and opportunity costs, *Getting to Neutral* estimated that natural solutions could store up to 25.5 Mt CO_2e/yr by 2045.

Pillar 2: Waste Biomass to Fuel

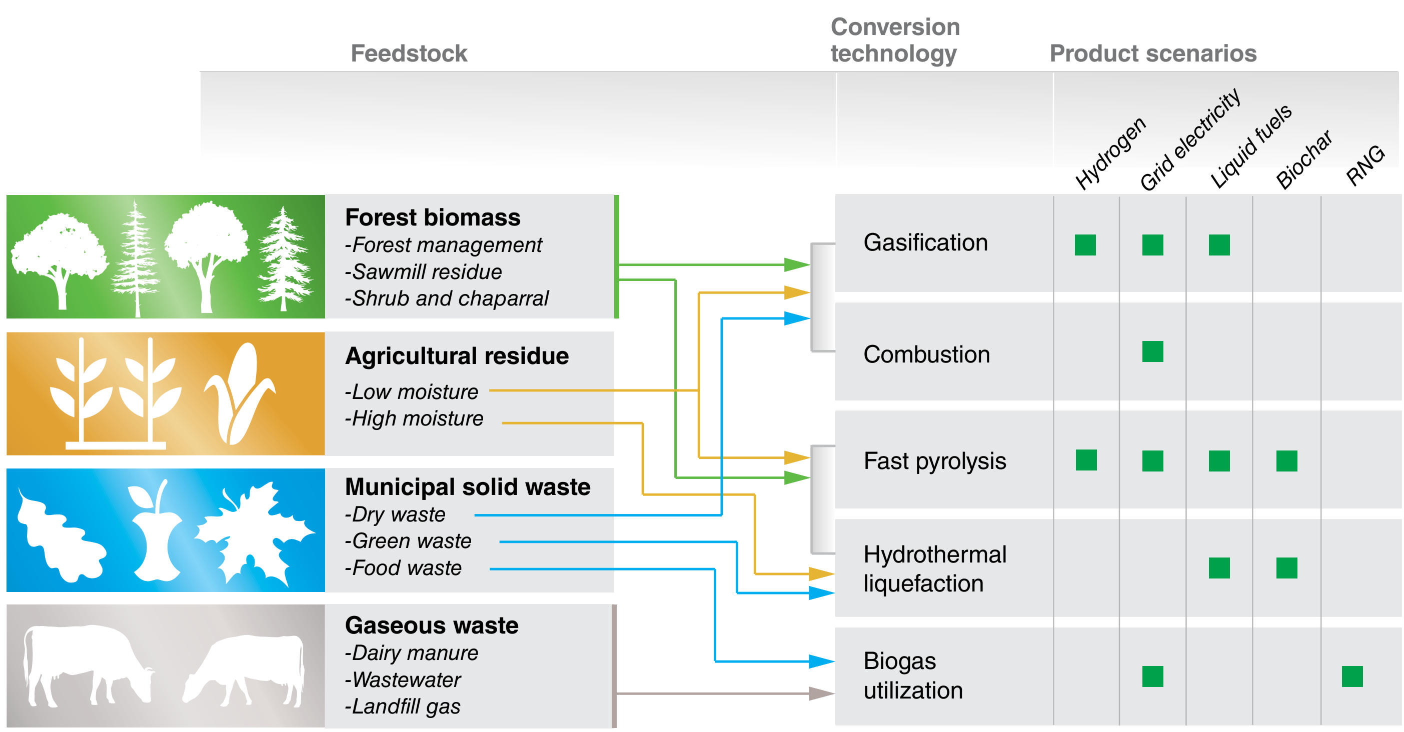
California has an estimated 54 to 56 million bone-dry metric tons (BDT) per year of available waste biomass such as agricultural wastes and residues (e.g., nut shells, rice hulls, and trimmings from orchards); landfill biogas; municipal solid waste; and sawmill waste. Forest residues—most importantly, woody debris removed to reduce the threat of fire in mature forests—are included in waste biomass as well. Today, waste biomass returns carbon to the atmosphere when it decays or burns in wildfires. Conversion strategies would transform California's waste biomass into fuels including liquid aviation fuels, renewable natural gas, and hydrogen (H_2), while capturing the CO_2 emitted during the conversion process. Captured CO_2 can be injected deep underground where it behaves like a liquid under high pressure, occupying microscopic pore spaces such as those that once held oil and gas within rock.

The report examined three primary methods for converting this waste to fuels—gasification, pyrolysis, and combustion—and touched on other, smaller-scale technologies. Gasification converts a fuel (biomass) into a mixture of gases, mostly carbon monoxide, H_2, and CO_2, at high temperature (900 to 1,200°C) and pressure. The product, known as synthesis gas or syngas, serves as a feedstock for producing industrial chemicals and other fuels or is burned to produce heat and power. Pyrolysis, a thermochemical conversion process operating at lower temperatures (440 to 700°C) and pressures than gasification, can decompose biomass into gas, liquid, and solid products. Gasification and pyrolysis should not be confused with combustion, the burning of biomass to produce heat and electricity. All of these technologies require the addition of CO_2 capture to the conversion process to achieve negative emissions. Biomass gasification to produce H_2 provided the maximum negative emissions potential,

Nameer Baker, a postdoctoral researcher working on a joint University of California, Berkeley–Lawrence Livermore study holds the dried roots of a perennial switchgrass (*Panicum virgatum*) that can increase soil carbon, nutrients, and water availability.

83 million tons per year, as well as the lowest cost per ton CO_2 of biomass strategies, while aligning with the state's renewable hydrogen goals. "Looking solely at technologies within the state, and at least at the pilot-scale, the most important technology was gasification of waste biomass to hydrogen gas," says Baker. "With a large enough plant, the process generates revenue while putting CO_2 underground."

The team also examined smaller-scale technologies, such as capturing CO_2 from biogas (renewable natural gas produced



This chart links biomass types to specific conversion technologies that help achieve carbon balance. Products of conversion include a range of energy sources including hydrogen gas, electricity, liquid fuels, biochar, a form of black carbon that can be added to soil, and renewable natural gas (RNG).

from fermentation of organic wastes such as food waste, trash, and manure in low-oxygen environments) to produce carbon-negative renewable natural gas for power and vehicle fueling. While smaller in supply than the agricultural and woody wastes, biogas is already produced in hundreds of facilities across the state and renewable natural gas has high value and varied uses, so could provide an early proving ground for production of carbon-negative biofuels.

Translating the variety of waste biomass conversion technologies and biomass types available, plus their associated products, carbon-storage options, costs, and timelines into real projects on the ground in California will require navigating a complex array of permits and authorizations, as well as cooperation between local, state, and federal agencies, commercial interests, stakeholders, and communities where carbon storage facilities might be located.

The Laboratory's 2021 report, *Permitting Carbon Capture & Storage Projects in California,* helps map one aspect of the complex path. The 2021 report maps out the permits and authorizations plants would need to obtain and describes the processes involved. George Peridas, report author and director of the Carbon Management Partnerships in the Laboratory's Energy Program, says, "We found that existing regulations are rigorous and capable of protecting people and resources. While additional regulation and legislation could tie up loose ends and make broader deployment of these technologies more efficient, no major reforms are needed to take the first steps and deploy the first few projects." The study found that the state needs first and foremost to focus on coordination of the permitting process among local, state, and federal agencies to succeed.

Stakeholder buy-in will also be essential. "An implementation barrier we scientists underestimate is the amount of change this solution—carbon storage and conversion—requires," says Aines. "We must engage communities and learn what is important for them. This conversation has to happen now." Some stakeholder communities have expressed interest in these types of projects. For example, California's fire-ravaged counties have shown interest in forest biomass conversion to reduce fire hazards, and landowners are willing to make their lands available for underground storage.

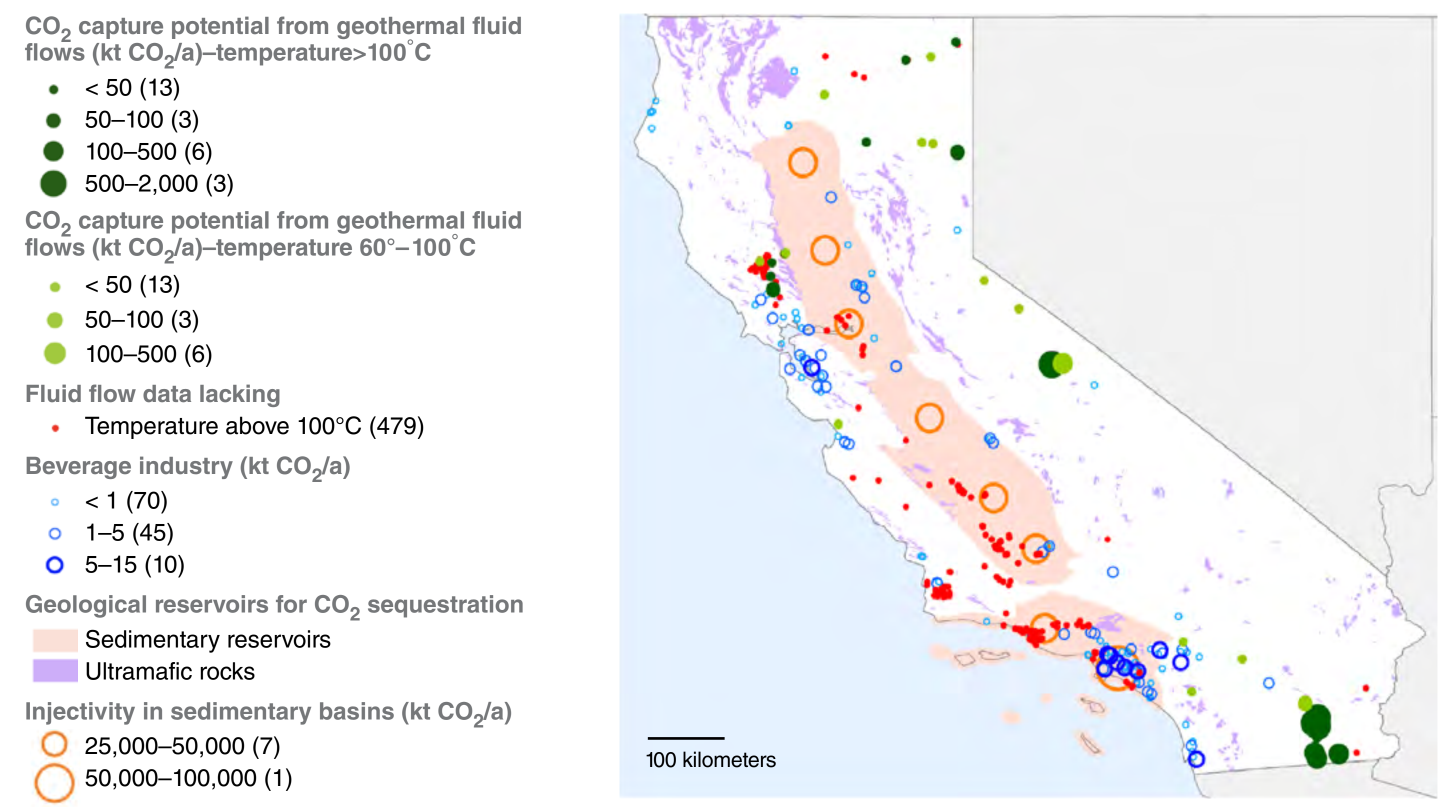
The report estimated that the total quantity of waste biomass available would be 56 million tons per year in 2045. If all of the carbon in this biomass were converted to CO_2 and sequestered, this total biomass resource would represent 106 million tons of CO_2, as an upper bound. The true amount of CO_2 that could be sequestered from waste biomass varies depending on the mix of conversion technologies adopted.

Pillar 3: CO_2 Capture with Machines

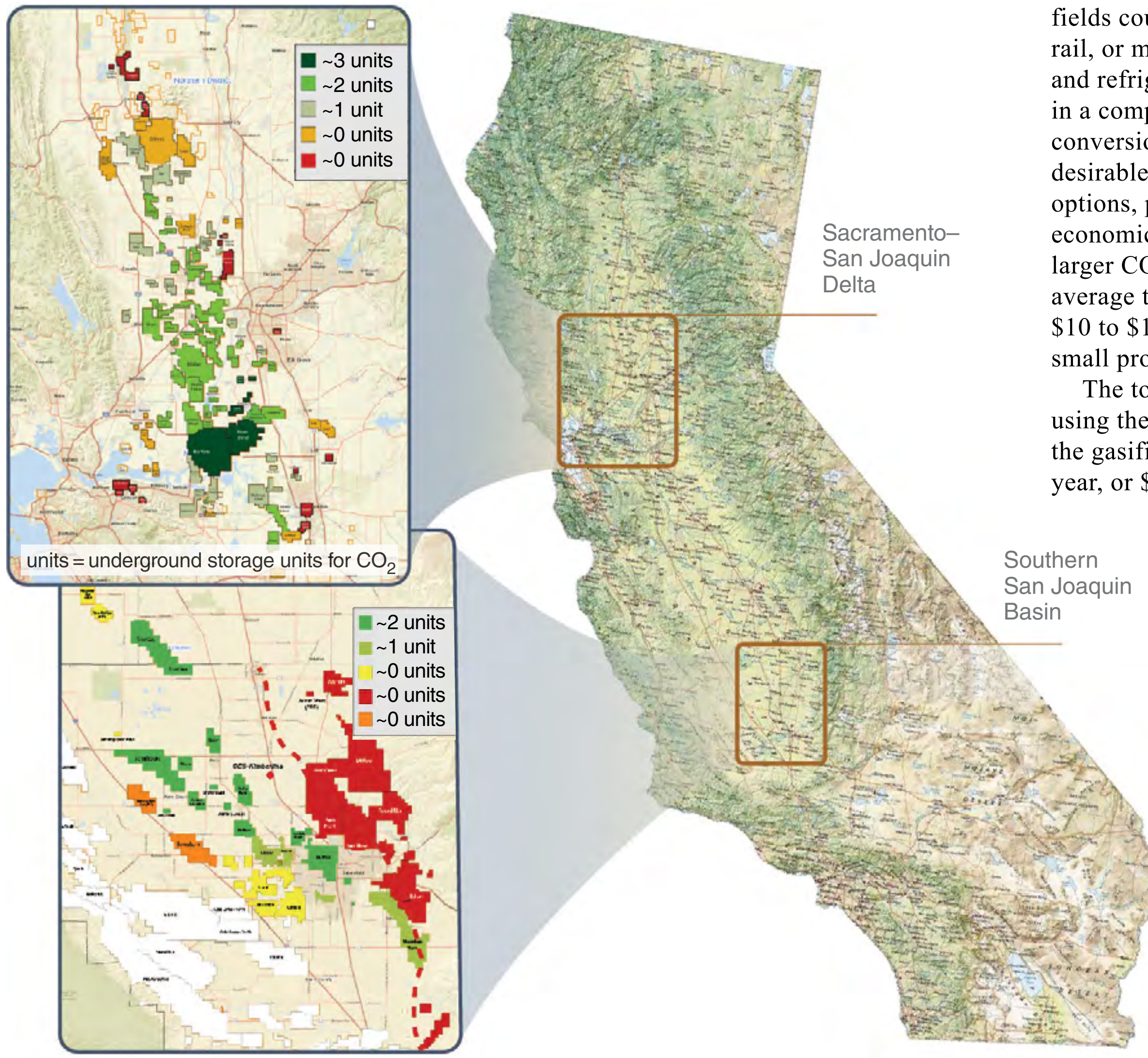
The previous two pillars offer inexpensive measures that have additional benefits beyond carbon capture, but those measures are not sufficient to meet the carbon reduction target. Purpose-built machines that absorb CO_2 into a solvent or onto a solid sorbent material, known as direct air capture (DAC), could provide the rest. The solvent or sorbent is heated to regenerate the material for reuse, simultaneously producing a concentrated stream of CO_2 for geologic storage. The process is energy intensive, requiring 180 to 310 megawatts of power for a CO_2 capture rate of 1 million tons per year.

"The large energy requirement means that we need to pair DAC technologies with low-carbon sources of energy, such as waste geothermal heat," says Simon Pang, a staff scientist in the Laboratory's Materials Sciences Division. In California, existing and potential geothermal energy from the Salton Sea, Mammoth Lakes area, and geothermal hotspots in northern parts of the state such as the Geysers could power as much as 11 million tons of sorbent-based DAC per year from new facilities, with 5 million tons supported by existing geothermal sources now used to generate electricity.

"Compared to other forms of carbon removal, the amount of CO_2 removed with DAC can be easy to measure and track," says Pang. "The downside is that DAC technologies are newer and relatively expensive." As more facilities are built, their cost will decline. Uncertainties in DAC technologies will be improved as companies get DAC facilities in the ground and operational to learn from and improve on the process. "The stability and lifetime of DAC materials is a major uncertainty," says Pang. "The sorbent material may degrade if it oxidizes in process operating conditions." Pang leads a Department of Energy (DOE) Office of Science, Basic Energy Sciences project at Livermore in collaboration with the National Renewable Energy Laboratory, Georgia Institute of Technology, and Global Thermostat to study how DAC sorbent materials degrade with the ultimate goal of developing materials with improved resistance to oxidation, leading to longer material lifetime.



Sources of CO_2 from California's geothermal energy fields and beverage (wine and beer) industries could be captured and sequestered. This map indicates areas with the potential to store CO_2 through direct air capture paired with geothermal energy to power the process. Light and dark green disks indicate geothermal activity. Orange circles show sites where CO_2 can be injected into sedimentary beds with available gas storage capacity. (kt CO_2/a = kilotons of CO_2 per acre.)



In two former oil fields identified for the underground storage of CO_2, areas shown in green are suitable for storage. Red indicates lack of suitability and other colors represent areas with insufficient data for analysis.

Storing CO_2 Underground

CO_2 captured with biomass conversion or DAC technology must then be stored underground. California's geology offers numerous areas with the potential to store carbon underground. Depleted oil or gas fields and deep saline formations containing brine provide sites to locate plants to store CO_2 in California's Central Valley. Livermore researchers used 3D geological models of two areas in the Central Valley—the Sacramento–San Joaquin Delta in the north, and the southern San Joaquin Basin in the south—to conservatively estimate their storage capacity at 17 billion tons, more than enough to meet the state's need.

Moving CO_2 from existing and potential biomass conversion and DAC sites and moving biogas fields to storage fields could be accomplished by truck, rail, or marine vessel in pressurized and refrigerated tanks or by pipeline in a compressed, dense state. Siting conversion plants near storage sites is desirable. Of all CO_2 transportation options, pipelines are the most economical for longer distances and larger CO_2 quantities. The system-wide average transport costs range from $10 to $18 per ton of CO_2 removed, a small proportion of the total system cost.

The total system cost of the scenario using the lowest-cost set of technologies, the gasification system, is $8 billion per year, or $65 per ton CO_2—a fraction of a percent of the state's gross state product. The biggest challenge to realizing the negative emissions vision of the *Getting to Neutral* report, according to Peridas, is the need to bring together stakeholders that have not worked together on one system."We need to have a wide range of experts from different fields such as oil and gas, the electric grid, and other technology sectors, farmers, regulators, communities, other stakeholders talking to each other," he says. "Each has different risk thresholds and priorities. The primary challenge is not technological. The challenge is the complexity of deploying infrastructure at scale."

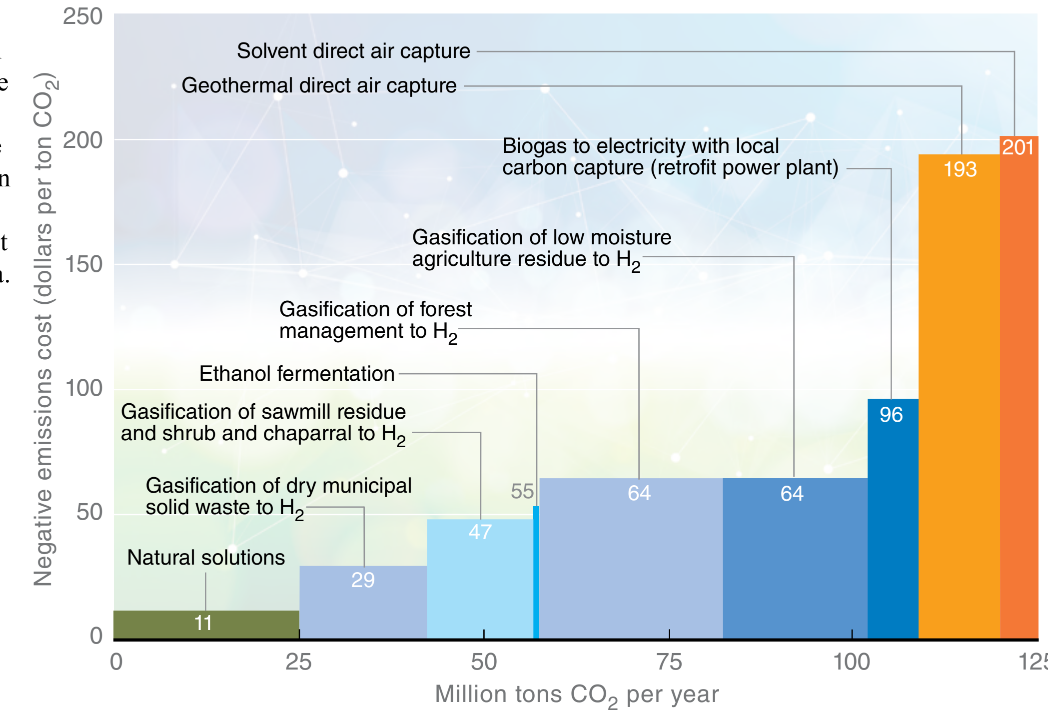
Another issue, according to Aines, is procuring long-term contracts for these plants: "The technology, raw materials, and state interest all exist," says Aines, "but it's easier to get a yearlong contract from funding agencies than a contract to supply one million tons per year of biomass for 20 years. Currently, no business performs the complete procedure of gathering biomass, processing it, storing carbon, and delivering the products. Geological storage of carbon and producing hydrogen have never been done before at this scale in the state." The report concludes that for the two locales examined in the Central Valley alone, the most conservative intermediate-resolution estimate yields capacities of about 17 billion tons, more than enough to meet the negative emission needs of California.

Plans for a Carbon Neutral Future

Laboratory scientists have briefed stakeholders from the California legislature and numerous state agencies to landowners, carbon storage technology companies, corporations interested in reducing their own carbon footprint, and academic researchers, as well as federal and other state and regional governments. Since *Getting to Neutral*'s release, state agencies have formulated plans to encourage the building of plants within the state to explore carbon storage strategies.

In February 2022, the Laboratory released a follow-on report titled "*Carbon Negative by 2030: CO_2 Removal Options for an Early Corporate Buyer*." Drawing on the expertise developed for *Getting to Neutral*, this report provides input to Microsoft Corporation's pathway to become carbon negative by 2030. Microsoft's goal will require the company to reduce its greenhouse gas emissions by over half and remove the rest, and then remove the equivalent of its historical emissions by 2050. The Livermore report will help guide Microsoft's pathway to carbon neutrality, but its recommendations apply to carbon markets globally, and to other corporations looking to accomplish the same thing. The Laboratory is also working on a report examining CO_2 removal potential at a county level, for the entire United States.

The released reports helped solidify the Laboratory's reputation as a leader in carbon removal research. *Getting to Neutral* won a DOE Secretary



A gasification scenario calculating potential negative emission pathways to carbon neutrality in California by 2045 prioritizes the production of hydrogen (H_2) over liquid fuel and electricity. Total (integrated) system cost for this scenario is $8.1B/year.

Achievement Award and a Livermore Laboratory Director's Science and Technology Award. The report helps inspire newly hired postdoctoral researchers and scientists to research pathways to negative emissions in such fields as soil science and carbon-conversion technologies. Lines of communication have opened to companies that offer DAC, pyrolysis, gasification, and anaerobic digestion technologies. The Laboratory has initiated projects to demonstrate some of the report's approaches on a small scale.

"We did something that I would not have imagined would happen with this report," says Aines. We created a map to carbon neutrality that also benefits communities, businesses, and the environment. We outlined what makes sense in California and simultaneously addressed the state's needs for carbon management, air pollution reduction, wildfire management, and stimulating the economy in the parts of the state that need it most. We mapped out where potential lies and how to make carbon neutrality happen. The phone started ringing. Now dozens of projects are in progress in California to implement what we outlined in the report."

—Allan Chen

Key Words: biochar, biogas, biomass, carbon dioxide (CO_2), carbon capture and storage, carbon neutral, ClimateWorks Foundation, combustion, direct air capture (DAC), gasification, geothermal energy, hydrogen (H_2), Laboratory Directed Research and Development Program, Livermore Lab Foundation (LLF), negative emissions, pyrolysis, renewable natural gas (RNG), soil carbon, syngas.

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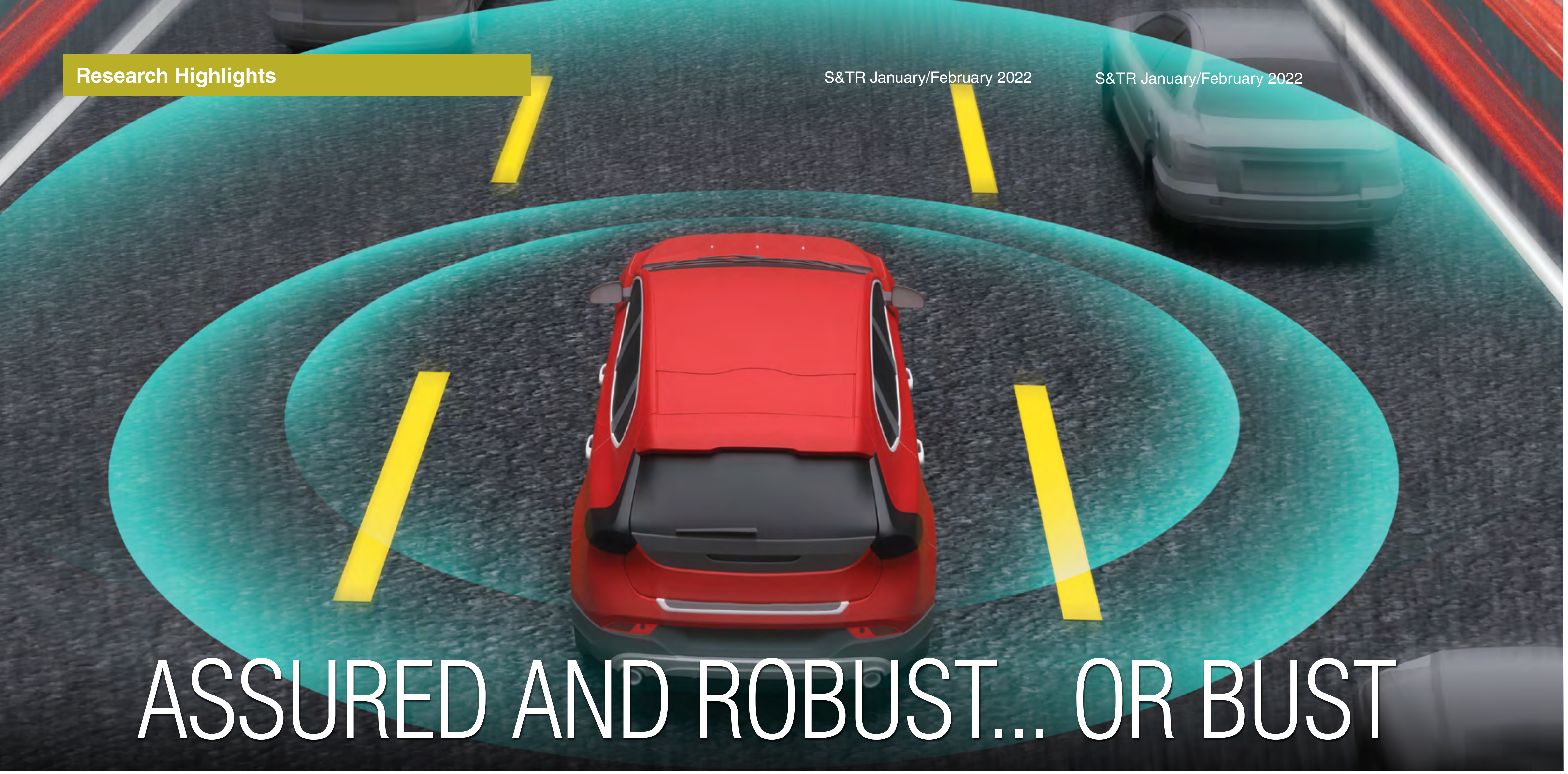
S&TR January/February 2022



ASSURED AND ROBUST... OR BUST

AN autonomous vehicle drives down a busy streeet when a strong breeze blows dust into the camera capturing the scene ahead and transmitting information to a machine-learning algorithm that detects potential safety hazards. In this hypothetical scenario, one misidentified object, such as an undetected person crossing the street, could mean the difference between life and death. Similarly, incorrect machine-learning predictions can incur significant costs in terms of experimental resources and lost opportunities in a range of scientific applications.

Many machine-learning systems employ complex mathematical concepts and algorithms to create artificial neural networks (ANNs) that mimic the biological neural network patterns of the brain, identifying patterns in data and then processing that information to make decisions and predictions. The algorithms are designed to identify critical differences among objects, but the rationale used by machines to make their predictions is opaque, as the systems teach themselves how to process the data within the networks. Even researchers designing and training the networks do not always understand how the models make decisions.

The consequences of a machine-learning error that presents irrelevant advertisements to a group of social media users may seem relatively minor. However, this opacity, combined with the fact that machine-learning systems are nascent and imperfect, makes trusting their accuracy difficult in mission-critical situations, such as recognizing life-or-death risks to military personnel or advancing materials science for Livermore's stockpile stewardship mission, inertial confinement fusion experiments, radiation detectors, and advanced sensors. (See *S&TR,* July/August 2017, pp. 16–19.) While opacity remains a challenge, Livermore's machine-learning experts aim to provide assurances on performance and enable trust in machine-learning technology through innovative validation and verification techniques. (See *S&TR*, March 2019, pp. 4–11.)

Robust at the Boundaries

The algorithms used in machine learning have achieved excellent performance on specific tasks such as image classification—distinguishing cars from trucks or cats from dogs—when training data is plentiful. Researchers have found that ANNs perform extremely well on clean, well-curated data that can be controlled; but when tested against real-world scenarios, where data is often messy, incomplete, or unpredictable, the ANNs fare much worse. Recent work has shown the algorithms remain extremely brittle, failing catastrophically in the presence of variations such as small rotations of an image.

"The lack of trust and transparency in current machine-learning algorithms prevents more widespread use in important applications such as robotic autonomous systems, in which artificial intelligence algorithms enable networks to gather information and make decisions in dynamic environments without human intervention," says Ryan Goldhahn, a computational engineer at Livermore. "To be truly useful, machine-learning algorithms must be robust, reliable, and able to run on small, low-power hardware that can perform dependably in the field."

Robust and assured machine learning implies that a machine-learning model makes a correct prediction under different perturbations (changes to the data) and noise (corruptions to the data). Robustness protects against both natural shifts in the real environment and adversarial shifts, such as a defaced traffic sign or data poisoning (intentionally altered data, imperceptible to humans, that is designed to fool the machine-learning model). Achieving robustness is a notable challenge.

Preparing for the Unexpected: Red AI

A team of Livermore researchers is developing "foolproof" or "assured" artificial intelligence (AI) to solve this critical concern. The team strives to identify the types of real-world perturbations (natural and adversarial) that might impact high-regret applications, in which consequences to human life are severe if the model makes a mistake. In a task once thought impossible due to the high number of inputs and complexity of nonlinear, deep neural networks, the team trains machine-learning models to provide guarantees—making them provably robust—by testing each input point in a computationally efficient manner. "Imagine being in a self-driving car that is prone to accidents when the weather changes," says Livermore computer scientist Bhavya Kailkhura. "With provable robustness guarantees, machine learning–AI can transfer control back to the human operator or make risk-averse decisions such as reducing speed when operating conditions such as heavy rain cross the boundary of a parameter."

To avoid catastrophic failures in mission-critical spaces, Livermore approaches improvements to robustness from two angles. Kailkhura calls the first angle the "Red AI" team, which deliberately fools machine-learning systems to anticipate where they will fail and then learn how to account for these failures in future, real-world scenarios. The Red AI team has explored training data poisoning, which degrades the robustness guarantees of state-of-the-art (SOTA) classifiers (decision-making tools used by AI systems) already used in machine-learning systems. To make the poisoning attacks used to train and test robust systems harder to detect, the Livermore team applies changes to the training data that are imperceptible to current AI systems, finding they can significantly reduce the robustness of SOTA classifiers and render existing defense approaches practically useless.

To better understand how the perturbations and noise work in real-world applications, Livermore has developed a system that deliberately corrupts image classifiers or detectors. In one approach, synthetic data is leveraged. "Tools from the video game industry allow us to render photo-realistic scenes and control the environment completely, altering background, objects present, the angle of the sun, and other features," says Goldhahn. The software, created to generate different images for training and testing, leverages 3D simulations to produce accurate, unique, and interesting data difficult to gather, curate, and label in the real world.

Foolproof (Assured) AI: Blue AI

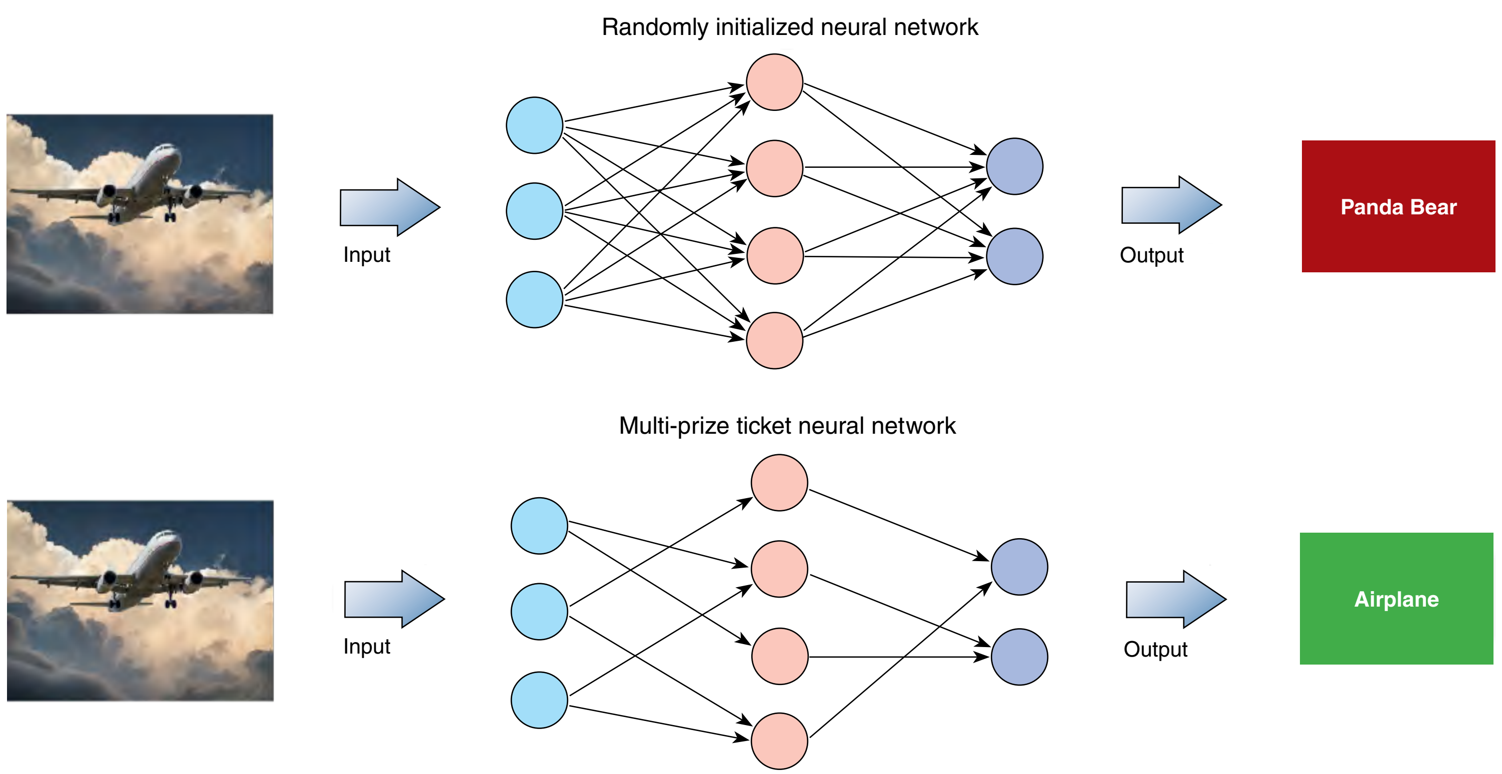
A "Blue AI" team complements the Red AI team's attacks by developing certified defenses that cannot be broken, regardless of the attacking algorithm, to provide robustness guarantees. Predictions from the certified model during testing are accompanied by a mathematical radius in which the predictions of the classifier are guaranteed to remain constant, thereby making them resilient to adversarial attacks on the system. If the input falls outside the mathematical radius under these circumstances, the predictions are still resilient.

Borrowing motivations from formal verification (computer-supported mathematical analysis ensuring software correctness), the Blue AI team developed certification schemes to provide rigorous guarantees on the correctness and robustness of pretrained machine-learning models under a range of natural and adversarial data shifts. Next, they developed training schemes to design machine-learning systems that are not only accurate but also provably correct and robust to multiple real-world perturbations such as shifting or rotating images, blurring, or altering lighting and contrast. "For the first time, the Livermore team has developed a mathematical tool that can provide decision makers with assurances about robustness to large, natural perturbations that can happen in the real world," says Kailkhura. "This tool essentially guarantees that the AI will do the right thing and make the right predictions under numerous specifications and constraints."

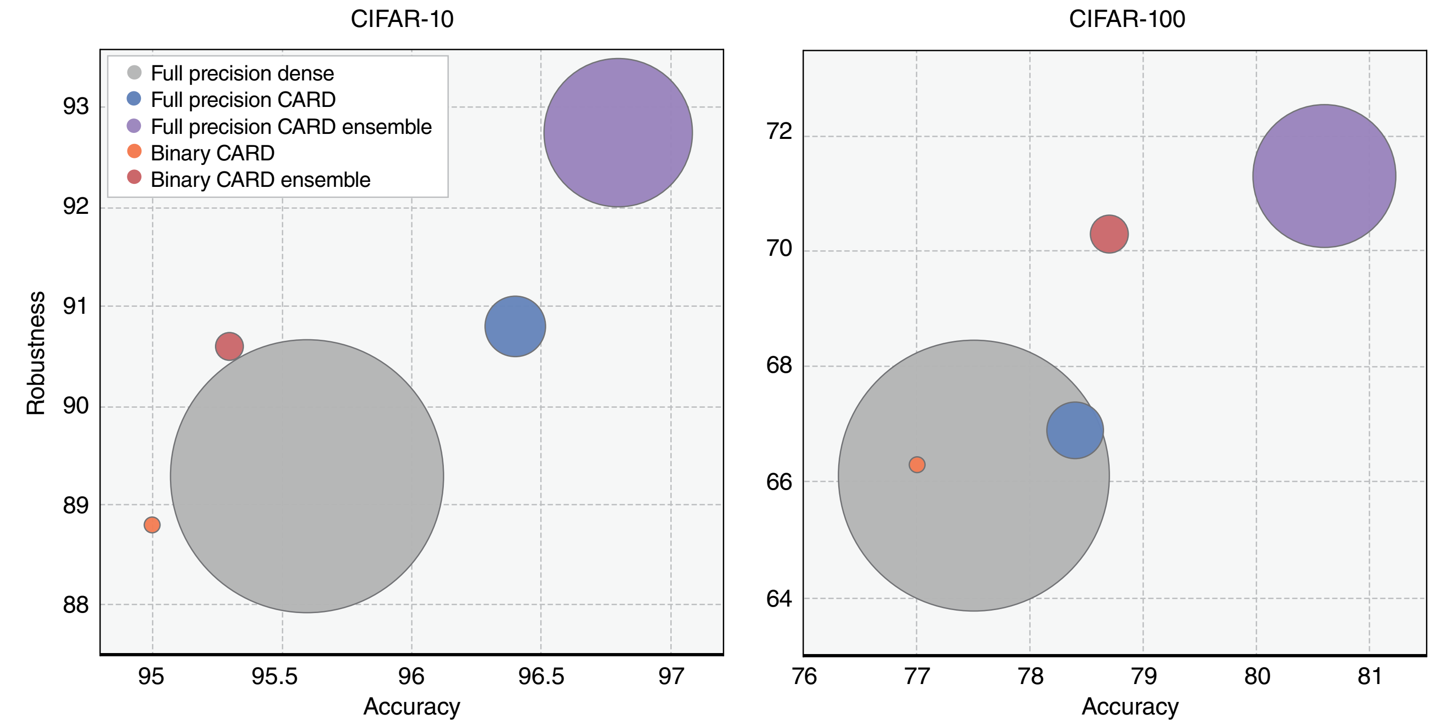
Real-World Deployment

The prevailing belief among researchers had been that only extremely large, dense neural networks can be robust, creating a challenge for running machine-learning systems on small, resource-constrained devices such as drones, smartphones, and other edge devices. The Livermore team has overcome this challenge with a multi-prize ticket approach creating models Kailkhura describes as "compact, accurate, and robust deep neural networks (CARDs)."

CARDS, sparse binary subnetworks, exist within a random-weight neural network in which weights are the model parameters that determine the influence of inputs on outputs. Despite having untrained weights, CARDS are drastically more robust compared to their more computationally expensive, traditional counterparts. "The math indicates, in principle, that you can make networks smaller while matching the accuracy and robustness of larger models," says Livermore postdoctoral researcher Brian Bartoldson. "This finding stands in sharp contrast to the current trend of making models bigger to achieve robustness."



Livermore's computational researchers have trained the multi-prize ticket neural networks to accurately classify information using fewer nodes than a randomly initialized neural network to make the right decisions in high-regret situations, despite perturbations such as blur and brightness.



As illustrated for two data sets (CIFAR-10 and CIFAR-100), certain model compression strategies can produce compact, accurate, and robust deep neural networks (CARDs) to provide comparable (or better) accuracy and robustness than the dense baseline while significantly reducing the memory overhead.

CARDs have important implications for Livermore's mission-critical space by enhancing the accuracy and reliability of machine learning for national security applications. Opportunities for commercial and research uses have emerged as well; CARDs enable accurate and robust AI models on computing platforms that had been too small to run large neural networks and that lacked the power to process huge networks on the devices themselves. Livermore computer scientist James Diffenderfer says, "If we could equip edge devices with their own neural networks or deep-learning models to classify the data they're collecting, we would accelerate discovery of what edge devices could do." In addition to everyday commercial uses—smartphones, watches, and other hardware—CARDs can benefit research areas such as space exploration that require equipment with small, efficient AI systems and models providing accurate predictions for months or years with few interventions.

These breakthroughs offer a huge opportunity to rapidly accelerate new discoveries in a variety of scientific fields and offer the potential to be much more accessible to institutions and research teams that lack the storage capacity, power, and high-performance computing requirements that large-scale machine-learning models require. Kailkhura says, "CARDs are significantly lighter, faster, and more power-efficient while maintaining state-of-the-art performance. They can unlock a range of potential uses in which deep learning struggles due to its inefficiency and brittleness. Collaborative autonomy; natural-language-processing models; and environmental, ocean, or urban monitoring using edge computing applications with affordable sensors are just some of the applications that could be better enabled by this research."

— Sheridan Hyland

Key Words: artificial intelligence (AI); artificial neural network (ANN); compact, accurate, and robust deep neural networks (CARDs); machine learning; multi-prize ticket; neural network.

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A SHOT LIKE NO OTHER

Researchers used a Laboratory-developed experimental platform at the National Ignition Facility (NIF) for the first-ever high-explosives shot at the facility. (Photo by Jason Laurea.)

HOW can researchers know what happens within the first few nanoseconds of a chemical reaction? Computational tools offer predictions about reaction behaviors. However, slowing time to watch a high-explosive reaction nanosecond by nanosecond and measure the evolution and phase change of solid reaction products is not possible.

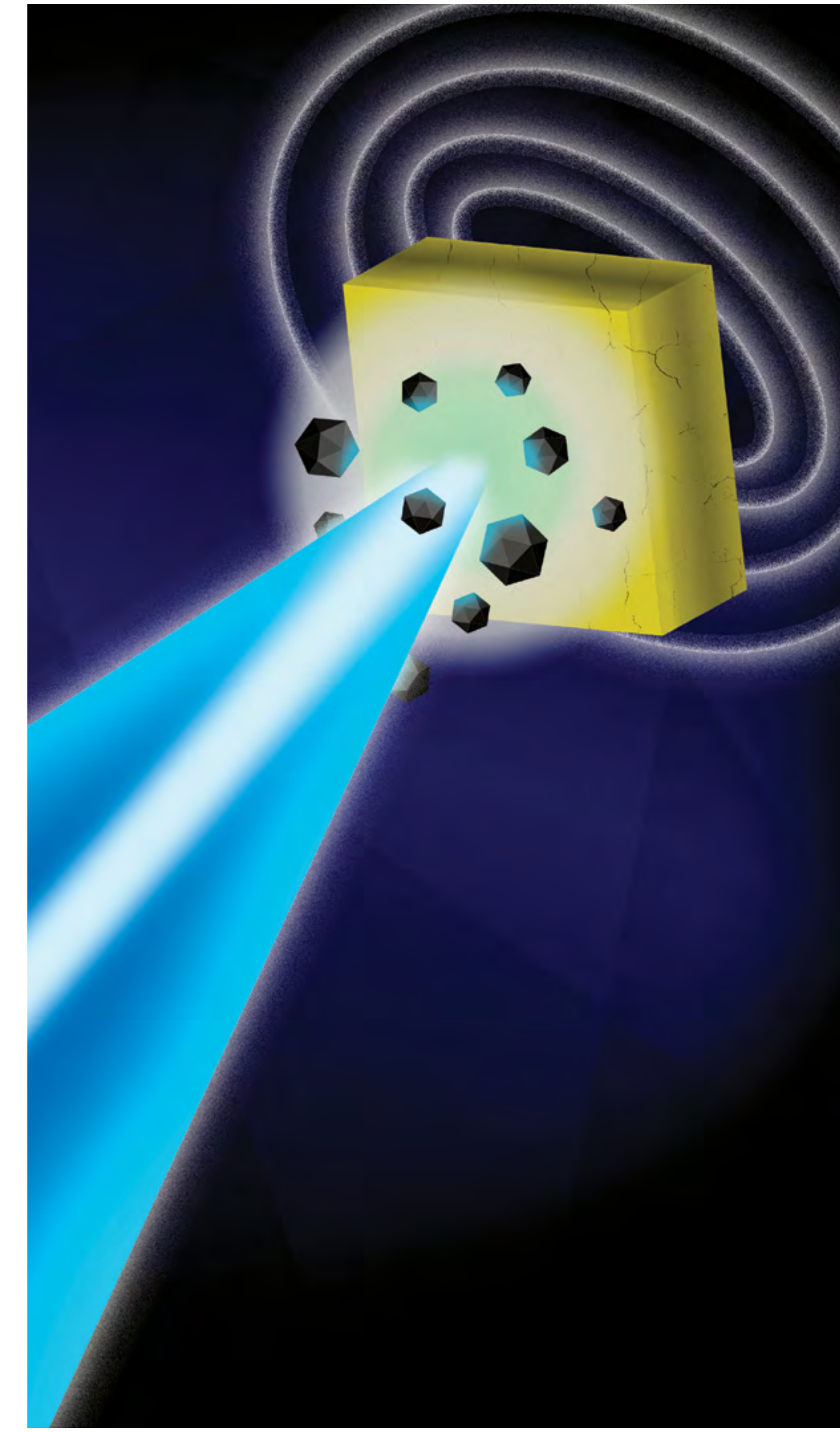
To address this scientific challenge, Lawrence Livermore National Laboratory researchers aligned energetic materials knowledge and high-energy-density science expertise with experimental capabilities at the world's most energetic laser, the National Ignition Facility (NIF). Directing NIF's long-laser drive and two x-ray probe beams at a non-detonable sample of high explosive provided data detailing the chemical evolution of a laser-driven high explosive over less than 50 nanoseconds. Precise diagnostics developed by Livermore researchers captured, for the first time, temporally and spatially resolved data to study the formation of solid carbon products (condensates) produced by the reaction and confirm their atomic structure. "The successful shot demonstrated the diagnostics are capable of characterizing the chemistry and pressure states of high explosives, in situ, during a reaction," says Samantha Clarke, lead scientist for the NIF experiment. "The team could not be more pleased with the outcome."

Until this experiment, the atomic structure of condensates produced by this type of high-explosive reaction had only been analyzed postdetonation or calculated by Livermore's physics and thermochemistry codes. "The experimental data, in turn, validates and improves predictive models such as Livermore's Cheetah thermochemical code and other hydrodynamic codes, improving future explosive simulations and analyses, predicting energy delivery for high explosives," says Larry Fried, one of the developers for Cheetah.

By helping scientists better understand and predict high explosives' behavior, the experiment and follow-on efforts will inform stockpile stewardship activities as well as nuclear detection and interdiction technologies supporting the Laboratory's counterterrorism and nonproliferation missions. "In addition to highlighting our capabilities and gaining unprecedented data, our success amplifies Livermore's leadership as a National Nuclear Security Agency Center of Excellence in High Explosives and Laboratory expertise in advanced diagnostics," says Lara Leininger, director of the Laboratory's Energetic Materials Center.

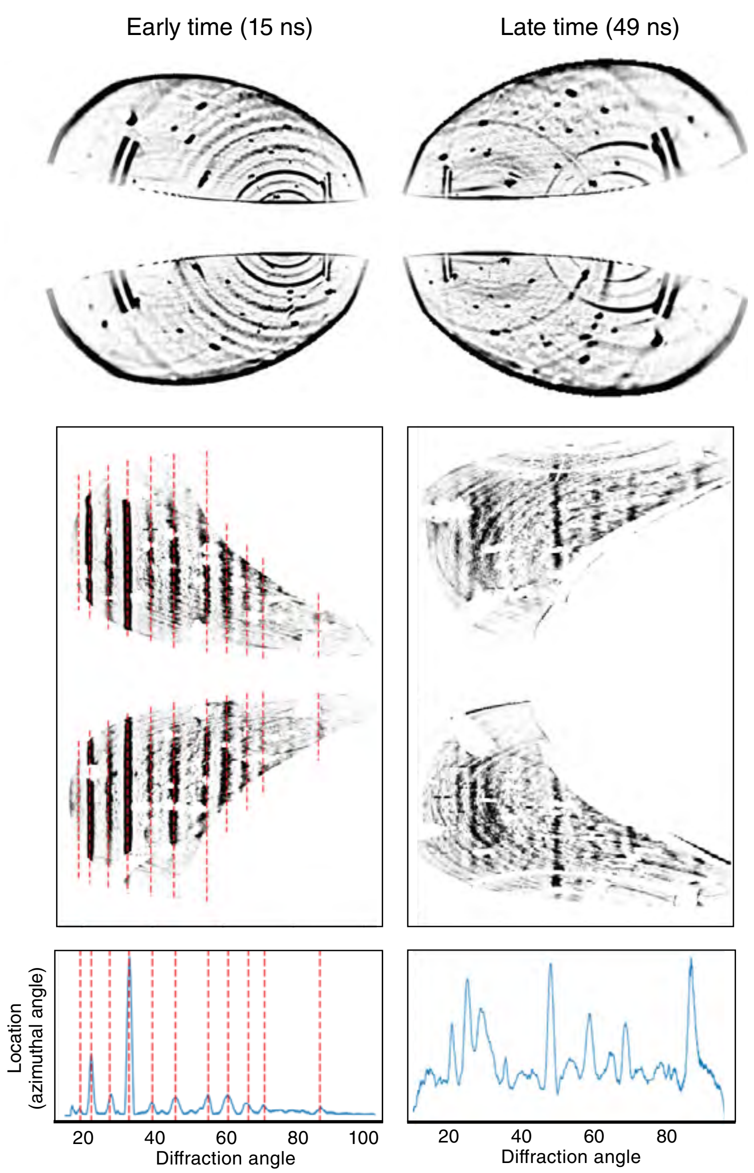
Three Years, Three Phases

The NIF experiment represented the final step in a three-year Laboratory Directed Research and Development (LDRD) Program project. A multidisciplinary team of physicists, experimentalists, and computational experts started work in 2017 to develop



An artist's rendering of the high-explosives (HE) shot at NIF depicts laser beams (blue stream) impacting an HE target (yellow square) and the resulting solid-carbon products (black polyhedrons). (Rendering by James Wickboldt.)

diagnostic techniques that, if successful, would provide temporally and spatially resolved data of a high-explosive reaction. The team leveraged techniques from earlier LDRD projects to grow the large, single crystals of explosive needed in initial tests of new



In diffraction data from the high-explosives shot at the early time point (left, XRS1 = 15 nanoseconds (ns)) and later time point (right, XRS = 49 ns), the Livermore team observed uncompressed triaminotrinitrobenzene (TATB) at early times (indicated by the dashed red lines) and evidence of reaction products, to be confirmed in future experiments, at the later time probe.

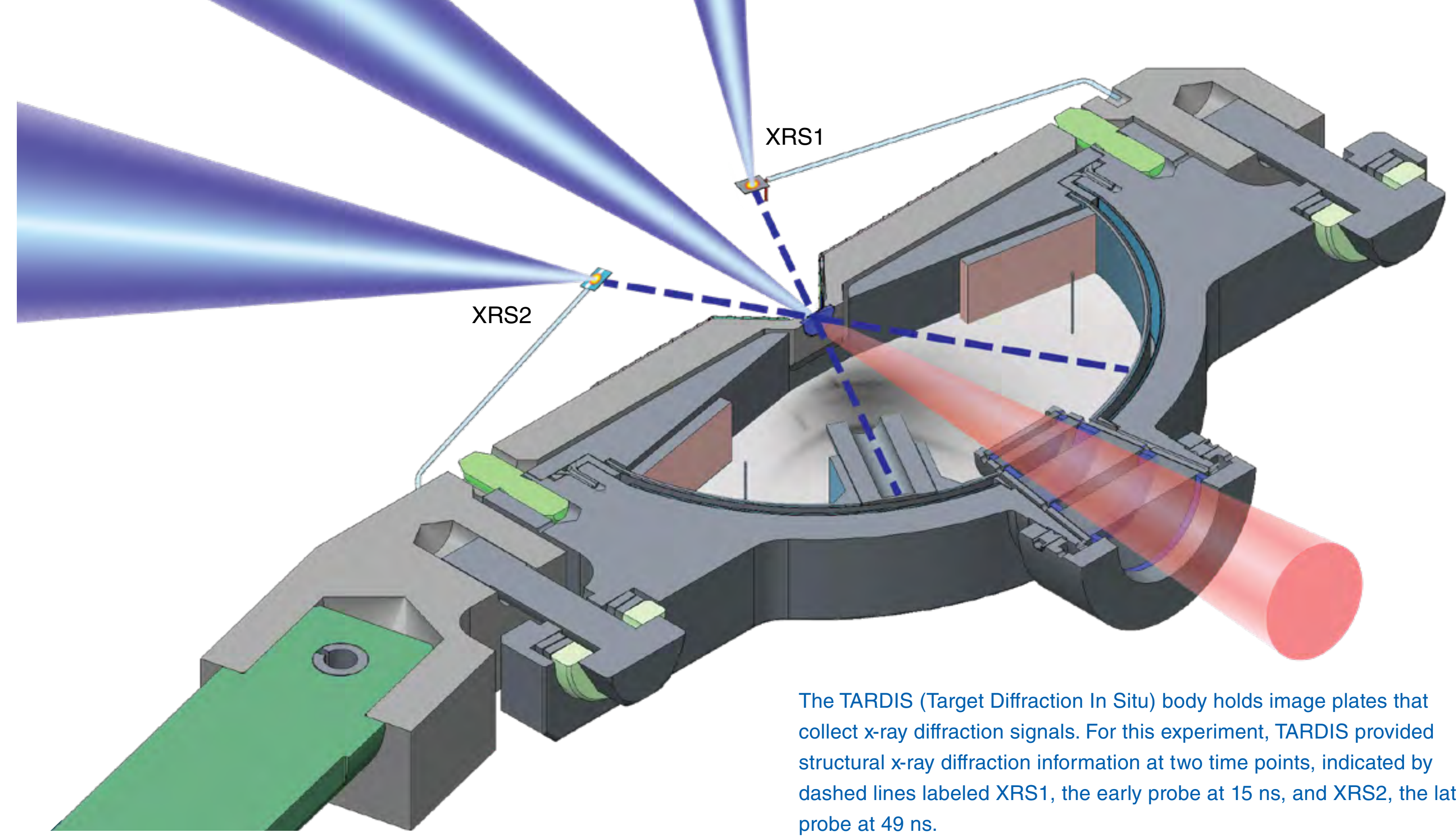
diagnostics and brought the project to scale based on experience with table-top, laser-driven experiments. "Projects with ambitious goals are based on many years of experience," says Leininger, the project's lead investigator.

Tests at the Laboratory's Jupiter Laser Facility ensured the new diagnostics could measure in situ, dynamic, laser-driven, high-explosive reactions. In the next phase, the team worked to ensure viability of the final NIF experiment. A series of shots at the Omega Laser Facility at the University of Rochester confirmed the experimental configuration, diagnostic setup, and the feasibility of measuring solid products in situ using x-ray diffraction with laser-backlighter x-ray probe beams.

In year three of the LDRD project, crystal preparation marked the home stretch on the path to an unprecedented NIF shot. "We decided to use thicker crystals to look at how the carbon products form in time," says physicist Ray Smith. "The thicker the sample, the greater the signal." A single crystal of the high explosive triaminotrinitrobenzene (TATB) in a non-detonable quantity comprised the target for the NIF shot. TATB offers low sensitivity to friction, pressure, temperature, impact, or spark, helping to ensure the safety of devices in the stockpile. Use of the explosive passed rigorous reviews by the Laboratory's Explosives Safety Committee and NIF Safety Program to ensure the safe execution of a high-explosive experiment. Says Ken Kasper, NIF Safety Program leader, "The advanced diagnostic systems were designed to extract experimental data from a small TATB sample. While considered thick for a crystal, the overall small sample size, less than 7 milligrams, made hazard management more straightforward."

The LDRD project encountered an unexpected obstacle when the COVID-19 pandemic moved the Laboratory into maximum telework mode, creating project delays. "The team had worked on this project for years and wanted to see it through," says Leininger. "Fabrication for the shot required months of preparation. Taking the shot too early would have been risky." Laboratory managers advocated for making the necessary capabilities available and extended the LDRD project for three months, giving the team time to test and obtain results. Teleconferencing platforms became more familiar as staff adjusted to teleworking. "With in-person activities scaled back, we relied on WebEx for close collaboration with NIF staff onsite and planned to run the experiment remotely," says Smith.

As the lead scientist, Clarke pushed ahead to coordinate all aspects of the experiment, working closely with lead designer for the shot, Damian Swift. Swift performed complex calculations to create a robust target design and laser drive to achieve the novel experiment's desired conditions. "My background was in diamond anvil experiments," says Clarke. "I quickly came up to speed with the skills and detailed design information required for this complex, laser-driven experiment."



The TARDIS (Target Diffraction In Situ) body holds image plates that collect x-ray diffraction signals. For this experiment, TARDIS provided structural x-ray diffraction information at two time points, indicated by dashed lines labeled XRS1, the early probe at 15 ns, and XRS2, the late probe at 49 ns.

A TARDIS Just in Time

Key to the success of the experiment was the application of the Laboratory-developed experimental platform known as TARDIS, for Target Diffraction In Situ. TARDIS integrates a NIF target and x-ray diffraction diagnostic on a single platform. "For this experiment, TARDIS would tell us what we wanted to know about the inside of the explosive as it detonates," says Fried. Although used for shorter duration experiments in the past, the team set out to apply TARDIS to a high-explosive shot with samples hundreds-of-micrometers thick and longer timescales than demonstrated previously at NIF. "We were expecting diamond formation to occur after significantly more than 20 nanoseconds, so we needed a longer drive and more time for the phase change to occur," says Fried. As a result, the experiment used stacked pulses on NIF, requiring the longest duration ablation (vaporizing material with the laser pulse to induce high pressure) attempted to date.

The team's goal was to apply a carefully designed laser pulse shape to shock-compress the sample to a steady pressure. Smith adds, "This duration of shock had never been done on TARDIS. As a compression wave propagates, the spot will get smaller and smaller, so we needed a larger initial laser spot." The team applied larger phase plates and special optical components to expand TARDIS's typical 1.8-millimeter-diameter spot to a 3-millimeter-diameter spot. Ultimately, TARDIS was prepared for the experiment's shock compression levels, which would exceed 150 gigapascals (1.5 million times the pressure of Earth's atmosphere).

While the three-year project closed with the history-making shot, the successful project is just the first step in a new series of experiments to improve understanding of high explosives' behavior. This work is continuing under the Dynamic Material Properties Campaign 2 funding, NA113. The improved diagnostics developed for the first shot will support Livermore research investigating the structure of products during high-explosive detonation and will validate predictive models. "We're continuing work to understand the data collected in this shot, continuing to interpret the lines and structures revealed," says Fried. "More experiments are needed for us to fully understand the chemical process behind high-explosive energy release."

A series of follow-on experiments evaluating a range of ignition levels from low-pressure to overdriven will further expand the Laboratory's understanding of high explosives. "We can nudge TATB to shock pressures beyond detonation conditions, speeding the dynamics to observe nanodiamond," says Cheetah developer Sorin Bastea. "Or we can keep compressing TATB to even higher pressures until we get no condensed carbon products at all. Each experiment helps improve and further validate our models." Leininger adds, "This work is not 'one and done.' Gathering more data and building our understanding will mean a great deal for chemistry and physics."

— Suzanne Storar

Key Words: Cheetah, energetic materials, Energetic Materials Center, high-explosive reaction, Laboratory Directed Research and Development (LDRD) Program, National Ignition Facility (NIF), TARDIS (Target Diffraction In Situ), target, thermochemical code, triaminotrinitrobenzene (TATB).

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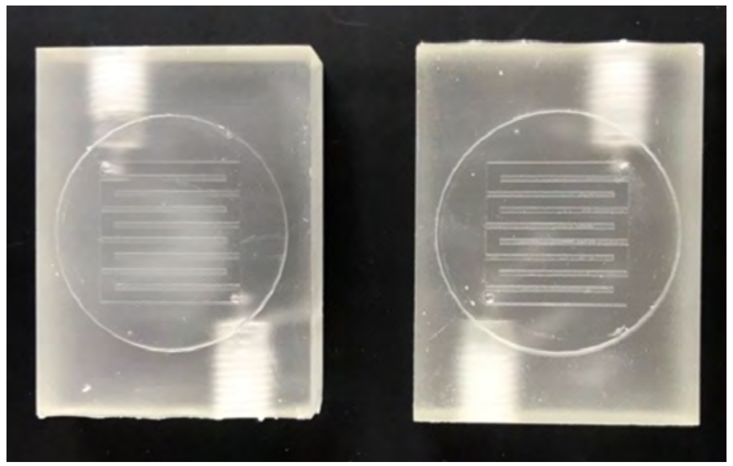
Research Highlights

ACCELERATING NUCLEAR FORENSICS IN THE FIELD

Nuclear and radiochemistry researcher Kevin Glennon holds the prototype forensic device. (Photo by Garry McLeod.)

FOUR critical questions arise immediately after a nuclear incident: What type of radioactive material was used? Where did the material originate? Could more devices or materials be found at large? And who bears responsibility? To answer these questions, nuclear forensics experts take field samples then transport them for laboratory analyses—a time-consuming method when, in the case of a catastrophic nuclear incident, time is of the essence. Recognizing the importance of faster analytical capabilities that can be conducted in real time at an incident site, a Laboratory Directed Research and Development (LDRD) Program–funded project led by radiochemist Narek Gharibyan led to development of a prototype chemistry-on-a-chip platform for field-deployable nuclear forensics. The assessment tool is designed to qualitatively identify actinides, specifically radioactive elements such as uranium or plutonium, sampled from postdetonation nuclear debris.

“Our approach allows us to use a very small sample and solution amount to quickly perform in-field analytical measurements of great forensic importance that would otherwise require a radiochemistry laboratory to analyze the samples,” says Gharibyan. Once the new platform is assembled, scientists inject a radioactive sample, and the device selectively filters the sample using a chemical extractant to separate the element of interest from the rest of the sample. After separation, scientists can quantify isotopic ratios in the separated element to determine the sample’s characteristics.



The chemistry-on-a-chip platform consists of two matching rectangular half-channels, 30 by 40 millimeters each, that, when stacked together, enclose a circular porous membrane 25 millimeters in diameter, permitting the extraction and separation of a desired material.

This method of extracting a radioactive element from a debris sample is comparable to preparing a cup of coffee, according to Gharibyan. "When brewing coffee, the caffeine is extracted from the grounds using hot water—the extractant—passing through a porous filter," he says. "Imagine an additional step in which caffeine content is isolated and quantified. Our chemistry-on-a-chip platform handles all these steps in one operation."

Streamlined Design

The chemistry-on-a-chip platform was created on 3D resin-based printers in the Livermore team's laboratory, giving researchers the flexibility to customize the module's design to the chemistry performed and to rapidly build and test the device using inexpensive, commercially available materials. The cost-effective design also made the platform practical for one-time use, avoiding any cross-contamination issues from sample to sample.

A flat-sheet supported liquid membrane (FS-SLM) modular design enables microfluidic capabilities similar to those in standard medical diagnostic systems. Anna Servis, a team member formerly at Livermore and now at Argonne National Laboratory, says, "To develop our chemistry-on-a-chip platform, we took the idea of a continuous-flow supported liquid membrane unit, a technology used in industrial hydrometallurgical processing, and scaled it down to process microscale nuclear samples." The platform requires only a submilliliter microsample, yielding easier, safer analyses for scientists to perform. Additional benefits of the microfluidic platform include rapid separation times, system portability, compact equipment setup, and the potential for automated separations with minimal human intervention.

Livermore's chemistry-on-a-chip nuclear forensic platform consists of two matching rectangular chips, 30 millimeters (mm) by 40 millimeters, with the bottom chip referred to as the "feed" and the top chip referred to as the "strip." Each chip has a raised cylindrical element that fits inside a matching inset in the other half. Once clamped together, the two chips align and enclose a 25-mm diameter porous membrane with channels for fluid transport, creating a three-tiered mechanism through which actinides can flow. The porous membrane acts as a filter, enabling the chemical separation process of one material or substance from another. Specialized aqueous solutions added to each half of the module initiate the flow of uranium from the feed to the strip by a combination of advection (transport by bulk motion of fluid) and diffusion (movement from higher to lower concentrations). After the device performs the necessary chemistry, the chip is taken out and discarded.

Optimized Flow

The team used previously acquired samples and synthetic debris to test the prototype and demonstrate how the platform would work in a real nuclear event. Computer-automated experiments controlled the quantity of solution injected and pushed through the module. Gharibyan's team performed measurements throughout the experimental runtime, tracking the amount of radioactive material transferred to the strip. On average, the device produced results in 20 to 30 minutes, with faster processing times for materials higher in concentration and slower times for materials lower in concentration.

The researchers designed and tested modules with four different channel depths—100, 200, 300, and 400 micrometers (μm)—to examine how channel depth affected fluid transport across the device. Optimal flow rates were found for the shallowest channel depth of 100 μm, achieving the team's goal to minimize the module's internal volume—requiring less solution—while maximizing the surface area available for mass transport across the enclosed membrane and increasing actinide extraction.

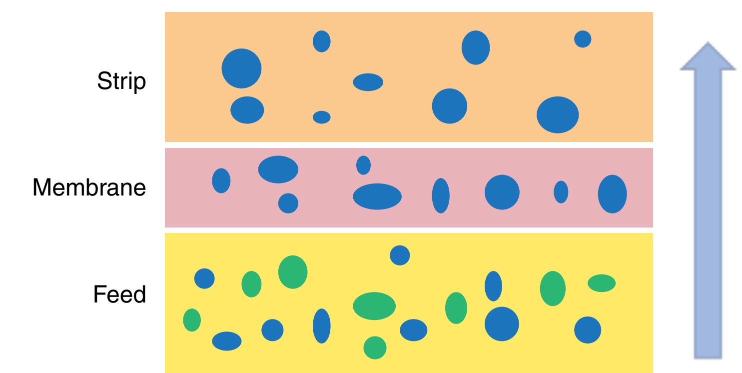
Gharibyan's team performed two kinds of separation experiments: single stage and multistage. For the single-stage separation of either uranium or plutonium, an extractant called tributyl phosphate (TBP) was added to the chip's membrane, causing the element of interest to bind to and join the extractant. TBP, coupled with the module's aqueous conditions, permitted the flow and transport of uranium and plutonium across the membrane and subsequently the strip, separating and purifying the sample. Once separated, a detector analyzed and quantified the stripped, "clean" sample. During a real nuclear incident, this analysis would tell scientists specific details about an element's characteristics and origin.

To separate a sample containing both uranium and plutonium, scientists used multistage separation methods and employed extra microfluidic modules for each additional separation step. First, the extractant TBP was added to the membrane, pulling uranium and plutonium away from other constituents in the sample. Since TBP extracts both uranium and plutonium, another extractant called tridodecylamine was added to pull plutonium away from uranium. The researchers repeated this second step to ensure the actinides were completely separated from each another. Once the two elements were separated and isolated on their own chip, a detector quantified each element's mass and isotopic ratios independently. Gharibyan says, "In both single- and multistage-separation methods, once the element is purified from all other material, the radioactive decay of the element's isotopes allows us to measure the amount of each radionuclide found, addressing parameters such as the age of the material, which can range anywhere from months to billions of years old."

After separation, the team, which included Laboratory postdoctoral researchers Kevin Glennon and Hector Valdovinos Uribe, quantified the stripped sample using a combination of nuclear and optical spectrometric techniques. Optical spectrometry uses ultraviolet–visible light and compares the sample to a blank solution for elemental quantification of total uranium content. The more light of a given wavelength that the sample absorbs, the greater the uranium concentration. Nuclear spectrometry analyzes alpha and gamma rays emitted from the collected sample to measure the content of uranium and plutonium radionuclides. The combined data from the optical and nuclear spectrometry measurements allowed researchers to determine the actinide ratios present in the samples.

The Next Chapter

LDRD sponsorship of the chemistry-on-a-chip platform was managed through the Laboratory's Physical and Life Sciences Directorate with funding from Global Security's (GS) Nuclear Threat Reduction Program at Livermore addressing nuclear counterterrorism, a core GS mission, and the need for fast-paced forensic analysis in the event of a nuclear catastrophe. With such a versatile platform, however, scientists can tailor the device for analytical measurements of actinides other than uranium and plutonium. Due to the separation capabilities of the device, a future application may include laboratory-based forensic techniques for purifying samples before they are injected into more complex instruments, so less material is needed to make high-quality measurements.



A radioactive sample is injected into the feed, and then the device's porous membrane selectively filters the sample using a chemical extractant. After 20 to 30 minutes, nearly all of the uranium (blue) has flowed to the strip to be quantified, filtering out any debris (green).

The technology also has the potential to measure fundamental extraction parameters of interest to heavy element chemists, addressing the difficulties in measuring short-lived heavy elements. "Microfluidic techniques are an underutilized tool with transformative potential in the field of inorganic chemical separations," says Servis. "Radiochemistry is a relatively small field. At the moment, few scientists experiment with 3D printing and microfluidics, opening a field full of opportunities."

Gharibyan's project has transitioned to a new initiative, the Post-Detonation Rapid Response Research (R^3) Venture, a multilaboratory collaboration supported by the U.S. Department of Energy's National Nuclear Security Administration Office of Defense Nuclear Nonproliferation Research and Development. The R^3 Venture microfluidics lead Jennifer Shusterman says, "Gharibyan and his team laid a strong foundation for the application of microfluidic devices in rapid, postdetonation debris chemistry. As a part of the new R^3 Venture, we hope to adapt the microfluidic platform to effectively measure actinides from more realistic debris samples."

— Shelby Conn

Key Words: actinides, chemistry-on-a-chip platform, elements, extraction, feed, flat-sheet supported liquid membrane (FS-SLM), isotopic ratio, Laboratory Directed Research and Development (LDRD) Program, microfluidics, multistage separation, nonproliferation, nuclear forensics, plutonium, porous membrane, postdetonation, radioactive, radiochemistry, single-stage separation, spectrometry, strip, uranium.

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Patents and Awards

In this section, we list recent patents issued to and awards received by Laboratory employees. Our goal is to showcase the distinguished scientific and technical achievements of our employees as well as to indicate the scale and scope of the work done at the Laboratory. For the full text of a patent, enter the seven- or eight-digit number in the search box at the U.S. Patent and Trademark Office's website (uspto.gov).

Patents

Solvent Depression of Transition Temperature to Selectively Stimulate Actuation of Shape Memory Polymer Foams
Anthony Boyle, Keith Hearon, Duncan J. Maitland, Landon D. Nash, Thomas S. Wilson
U. S. Patent 11,109,869 B2
September 7, 2021

Electromethanogenesis Reactor
Jennifer Marie Knipe, Sarah E. Baker, Marcus A. Worsley, Swetha Chandrasekaran
U. S. Patent 11,111,468 B2
September 7, 2021

Separation of a Target Substance from a Fluid or Mixture Using Encapsulated Sorbents
Roger D. Aines, Christopher M. Spadaccini, Joshuah K. Stolaroff, William L. Bourcier, Jennifer A. Lewis, Eric B. Duoss, John J. Vericella
U. S. Patent 11,117,091 B2
September 14, 2021

Optically Enhanced Patternable Photosensitivity via Oxygen Excitation
Ryan Hensleigh, Eric Duoss
U. S. Patent 11,117,361 B2
September 14, 2021

Mimicking of Corruption in Images
Rushil Anirudh, Peer-Timo Bremer, Jayaraman Thiagarajan, Bhavya Kailkhura
U. S. Patent 11,126,895 B2
September 21, 2021

Lattice Microfluidics
Joshua R. Deotte, Sarah Baker, Eric Duoss, Jennifer Marie Knipe, Fang Qian, Samantha Ruelas
U. S. Patent 11,130,131 B2
September 28, 2021

Dual Wavelength Negative Imaging DLP-SLA System
Bryan D. Moran
U. S. Patent 11,130,288 B2
September 28, 2021

Opto-Thermal Laser Detonator
Paul R. Wilkins
U. S. Patent 11,130,530 B2
September 28, 2021

Multi-Fluid, Earth Battery Energy Systems and Methods
Thomas A. Buscheck, Ravindra Shrikrishna Upadhye
U. S. Patent 11,137,169 B2
October 5, 2021

System and Method for Compact, Adaptive Optical Sensor for Obtaining Areal Images
Robert Matthew Panas, Harris J. Hall, Lavern A. Starman
U. S. Patent 11,137,591 B2
October 5, 2021

Incident Exercise in a Virtual Environment
Gregory K. White, William H. Dunlop, T.R. Koncher, Steve Kreek
U. S. Patent 11,138,902 B2
October 5, 2021

Shape-Memory Polymer Foam Device for Treating Aneurysms
Jason M. Orteha, William J. Benett, Ward Small, Thomas S. Wilson, Duncan J. Maitland, Jonathan Hartman
U. S. Patent 11,141,164 B2
October 12, 2021

Additive Manufacturing Powder Spreading Technology to Mitigate Surface Defects
Saad A. Khairallah, Gabe Guss, Eric B. Herbold, Wayne E. King, Manyalibo J. Matthews, Alexander M. Rubenchik, Otis R. Walton
U. S. Patent 11,141,912 B2
October 12, 2021

Awards

The **Fusion Power Associates Board of Directors** selected Livermore physicist **Tammy Ma** for its **2021 Engineering Award**. The award recognizes professionals with technical accomplishments and leadership potential in the fusion field. Ma has authored or co-authored more than 180 refereed journal publications in fusion energy and plasma research. She leads the High-Intensity High-Energy-Density Science Element in the Advanced Photon Technologies Program within the National Ignition Facility and Photon Science Principal Directorate, and she serves as co-deputy director for Livermore's Laboratory Directed Research and Development Program. Ma recently served on a Department of Energy subcommittee that authored a long-range strategic plan for fusion energy and plasma science.

The **Council of the American Meteorological Society** (AMS) awarded its **Henry G. Houghton Award** to Livermore atmospheric scientist **Mark Zelinka**, the task lead for Cloud Feedbacks in the Laboratory's Program for Climate Model Diagnosis and Intercomparison. The award recognizes early career scientists with research achievement in physical meteorology, physical climatology, atmospheric chemistry, or hydrology. AMS cited Zelinka for his work studying the planet-cooling effect of clouds to better understand climate sensitivity—global warming due to doubling of carbon dioxide. His research examines how changes in the balance of competing cooling–heating effects can intensify or dampen the climate's response to increasing greenhouse gases.

Abstract

The Path to a Carbon Neutral California

The Livermore report *Getting to Neutral: Options for Negative Carbon Emissions in California* outlines a strategy to reduce California's carbon emissions to net zero by 2045. The report demonstrates that the state could achieve its carbon neutrality goal at modest cost, using resources and jobs within the state and with technology that is already demonstrated or mature. Existing state plans chart a pathway to reducing 80 percent or more of those emissions. By applying findings in the *Getting to Neutral* report, California could gain the remaining 20 percent emissions reduction through negative emissions—carbon dioxide that is physically removed from the atmosphere through approaches such as biomass growth or direct air capture.

Contact: Sarah Baker (925) 422-3811 (baker74@llnl.gov).

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Coming Next Issue

Immune Infrastructure

The Laboratory is developing broad, integrated capabilities to increase the security and resilience of our critical infrastructure from nation-state cyberattacks.

Also in the next issue:

- *Innovative 3D nuclear battery prototypes developed at the Laboratory could potentially increase power density and duration for remote applications.*
- *GEOSX, an open-source, high-performance simulator for large-scale geological carbon dioxide storage, assists with carbon neutrality efforts.*
- *Laboratory recipients of the Department of Energy's Office of Science Early Career Research Program demonstrate exceptional promise.*

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