Renowned Accelerator Facility TURNS 30

Livermore's Center for Accelerator Mass Spectrometry celebrates a milestone anniversary with wide-ranging new research endeavors.

DURING autumn of 1989, scientists at Lawrence Livermore's Center for Accelerator Mass Spectrometry (CAMS) were mystified. Electrical systems connected to the center's premier accelerator system, the high-precision instrument at the heart of CAMS's radiocarbon work, were intermittently going offline, bringing experiments to frequent, frustrating halts. At the time, the Laboratory's reputation for Earth science and biomedical research was gaining momentum. Any hiccup in the center's ability to analyze samples from national laboratories, universities, and other institutions was unwelcome.

After some investigation, the source of the problem was identified—a power coil had shifted slightly, which in turn affected the electrical grounding within the accelerator. The culprit was the 6.9-magnitude Loma Prieta earthquake, whose epicenter had been just 96 kilometers south of the Laboratory. "Until then, none of us had needed to debug the equipment after an earthquake," remembers Jay Davis, CAMS's founding director. "We did not expect to have to troubleshoot that kind of problem." The response to the challenge was exemplary of the center's overarching methodology. Since CAMS opened in 1988, problem solving, insistence on accuracy, and adaptation to change have been integral to facility operations, including for experiments and technology development, and have helped make it a reliable facility for supporting Livermore's national security mission.

Similar to many other nascent efforts at the Laboratory, accelerator mass spectrometry (AMS) was initially used for nuclear technology research. (See *S&TR*, December 2013, pp. 16–19.) However, since Livermore's leadership in fundamental science was growing into more areas of physics, chemistry, and materials science, early plans for an AMS-focused facility concentrated on anticipating future needs and providing service to a wide community of user. CAMS originally supported only radiocarbon and tritium analysis—with separate handling protocols and preparation laboratories to avoid contamination. A major goal was to expand capabilities for measuring other isotopes. Davis says, "CAMS opened the door to exploration. Scientists started to ask themselves what could be done with this technology."

In the beginning, resources were modest. CAMS staff assembled the first accelerator by combining new and used equipment. Davis recalls, "The ion source was purchased new, but we harvested the magnets from other laboratories. We built a machine we could not afford to buy." The team set about constructing instrumentation safely and inexpensively with next-generation computer systems. "Measuring isotopes at 1 radioactive atom per 1 quadrillion atoms is difficult to achieve. One big challenge was setting up the computer-controlled instrumentation that makes these measurements possible," continues Davis.

CAMS's focus has been guided over three decades by attention to both applications and technical development. "We have achieved a balance between internal and external users and

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Livermore scientist Jessica Osuna (left) and former colleague Alexandra Hedgpeth monitor gas emissions from a soil column at a Minnesota peat bog. Sample analysis is done at Livermore's Center for Accelerator Mass Spectrometry (CAMS).

between fundamental basic science and Laboratory programmatic work," explains CAMS director Graham Bench. Today, the center houses multiple AMS instruments including the 10-megavolt FN tandem Van de Graaff accelerator, a heavy-element beamline for analysis of plutonium and uranium isotopes, a beamline for ion implantation experiments, and finely tuned devices for ion and neutron production. (See the box on p. 7.) A dedicated biological accelerator mass spectrometry (bioAMS) resource leverages high-performance liquid chromatography for specialized measurements. (See *S&TR*, December 2012, pp. 18–20.)

Built for Change

Although many similarly sized user facilities around the nation specialize in one research area, CAMS supports multidisciplinary goals and technologies with fewer than 30 full-time staff. Bench attributes the center's longevity to its scientific diversity. "We have always had a broad viewpoint. CAMS is unique because of our wide scope of work," he says. Ken Turteltaub, the Laboratory's Biosciences and Biotechnology Division lead, agrees, "We wanted to create an environment that brought different types of scientists together."

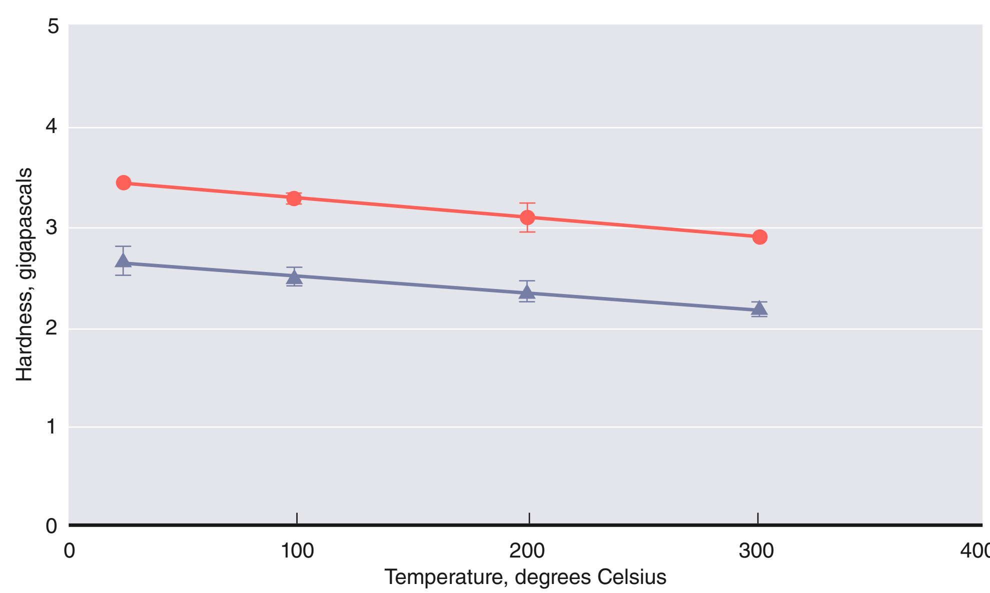
CAMS staff routinely measure a bevy of long-lived radioisotopes, including tritium; carbon-14 (^{14}C, in both liquid and solid forms); cosmogenic nuclides such as beryllium-10 (^{10}Be), aluminum-26 (^{26}Al), and chlorine-36 (^{36}Cl); nuclear fission decay products such as iodine-129 and strontium-90; and rare-earth metals and actinides including uranium-233 and -236 and plutonium-230 and -240. From studies of Earth system processes, atmospheric chemistry, and drug metabolism to assessments of radiation damage and materials modification, CAMS scientists have produced more than 1,000 peer-reviewed publications.

This range of expertise enables the center to support a broad user base of academic and commercial collaborators. (See the box on p. 10.) In addition, several U.S. agencies rely on CAMS, including the Department of Energy (DOE), the National Institutes of Health (NIH), and the National Science Foundation.

Building on CAMS's 30-year legacy of scientific achievements, the center's leaders emphasize the importance of persistent innovation and forward-looking research. "Our latest accomplishments are as important as what we have done in the past," states Bench. Davis adds, "CAMS was built for change." Recent work in four research areas showcases the breadth and depth of the center's capabilities: unique solutions for nuclear science challenges; studies of the terrestrial carbon cycle; geochronological and paleoclimatological research with cosmogenic nuclides; and laser-based ^{14}C analysis of biomedical samples.

Nuclear Science on a Mission

CAMS's highly sensitive instrumentation and nuclear science expertise are essential to Livermore's mission work. (See *S&TR*, September 2011, pp. 4–10.) Researchers such as deputy group leader Scott Tumey use high-energy ion implantation to study radiation damage. By accelerating ions into a target to change its properties, Tumey's team can study how tolerant materials are to radiation damage, and thus determine their utility in nuclear reactors. "The key to using ions for studying radiation damage is understanding the fundamental differences between how ions and neutrons interact with matter," explains Tumey. The higher the energy used in the irradiation process, the deeper the ions can penetrate the target material. For Tumey, a CAMS research scientist since 2004, this capability is invaluable. "We can produce samples on



A research team led by Livermore scientist Scott Tumey compared unirradiated (purple line) and ion-irradiated (pink line) steel alloys to characterize how indentation depth affects a material's hardness. The graph shows that at a depth of 200 nanometers, the effect was less pronounced after irradiation and at higher temperatures.

Safely Separating Ions and Counting Atoms

Lawrence Livermore's Center for Accelerator Mass Spectrometry (CAMS) occupies approximately 929 square meters and encompasses a dozen experimental and preparation laboratories. The FN tandem Van de Graaff accelerator is the center's principal instrument and can run a single radiocarbon sample in about 15 minutes. The 10-megavolt device provides high-precision, high-energy analyses of multiple isotopes with the sensitivity to detect 1 radioactive atom among 1 quadrillion atoms. Annually, CAMS's instruments average a combined 25,000 isotopic measurements.

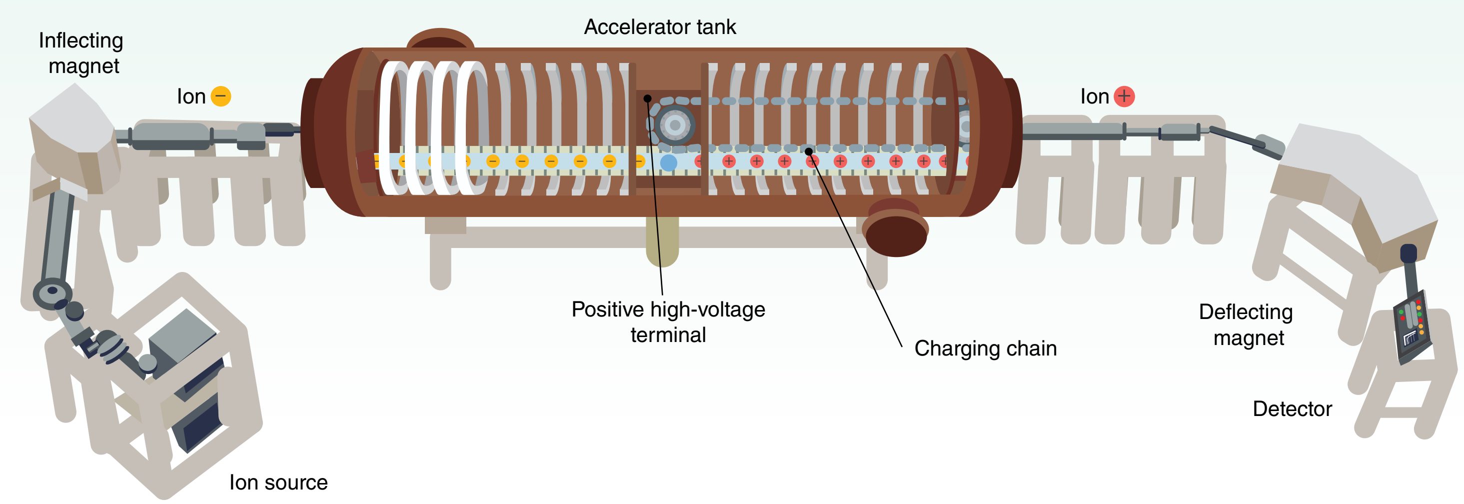
CAMS director Graham Bench stresses the importance of vigilance and safety in the center's research activities. "One can never be too careful when working with high-voltage machines and radioactive materials," he says. Tom Brown, CAMS's deputy director, ensures all new staff undergo safety training for using accelerator mass spectrometry (AMS). He notes, "High voltage, asphyxiating gas, and ionizing radiation are the main operational hazards. We follow several levels of controls to ensure personnel safety and environmental protection."

CAMS complies with both Lawrence Livermore's and the Department of Energy's safety procedures, and its laboratories are equipped with numerous safety features. Although most AMS operations do not produce substantial radiation, monitoring systems continuously run to enable automatic shutdown of a system if radiation levels exceed a certain limit. Power supplies are surrounded by grounded shields, and special sensors detect gas leaks and low oxygen levels. "We have never had a significant gas leak, nor have we needed the oxygen monitoring system to trigger for an actual dangerous event," emphasizes Brown, who joined CAMS 24 years ago.

With safety protocols in place, CAMS staff perform routine maintenance on all instrumentation. If a component malfunctions, Bench explains, "We fix it ourselves. We do as much as we can onsite." Replacement parts can be ordered from manufacturers, yet the CAMS team assumes responsibility for installation, configuration, and associated mechanical and computerized tasks. For instance, inside the FN's tank, acceleration is achieved by moving chains that maintain a constant positive charge on a high-voltage terminal. The chains' plastic and metal parts and the bearings around the chains' wheels can wear down and must be replaced. To access these components, the team must first pump sulfur hexafluoride—an insulating gas commonly used in high-voltage particle accelerators—into a storage container and then fill the tank with breathable air. According to Brown, staff develop do-it-yourself skills through their own experiences at CAMS and through dialogue with colleagues at other accelerator facilities.

High demand for CAMS's capabilities means accelerators must run all day and sometimes overnight, so the center's safety track record is crucial to its success. Brown says, "We all understand that we must control the hazards we encounter while conducting our work."

In a typical radiocarbon experiment, samples loaded into the accelerator's ion source are bombarded by negatively charged cesium ions to produce carbon ions. A magnet is set to select the atomic mass of interest. Inside the main tank, the particles are accelerated in two stages to separate the interfering molecular ions from their constituent atomic ions. A deflecting magnet then separates the ions by atomic mass and charge. A detector counts the ions of interest, and isotopic ratios are calculated to reveal the ions' relative abundance.



CAMS staff scientist Susan Zimmerman shows a core sample of mud retrieved from the bottom of Mono Lake. Radiocarbon dating of the sediments is done with the CAMS accelerator, and results help determine the environmental history of the lake basin.

which physical properties such as tensile strength and hardness can be directly measured rather than inferred from microstructural examination," he says.

In 2017, CAMS joined the Nuclear Science User Facilities network. Funded by DOE's Office of Nuclear Energy, this group of experimental laboratories provides facility access and technical proficiency to researchers tackling nuclear energy challenges. CAMS provides a unique opportunity to study the effects of neutrons on various nuclear materials, such as fuels. Tumey says, "With our high-energy accelerator, we can irradiate samples over a much larger range than other ion beam facilities, thus producing specimens suitable for the direct measurement of physical property changes associated with radiation damage."

In addition, AMS complements other analytical techniques to advance the Laboratory's nuclear forensic capabilities. "Our tandem accelerator effectively removes all molecules that have the same mass as the isotopes of interest. This feature provides an incredibly high degree of selectively and sensitivity," notes Tumey. For example, ^{26}Al and ^{10}Be can reveal information about materials used in a nuclear device, and thus Tumey's team is developing these isotopic signatures for post-detonation nuclear debris. He says, "Our methods are robust and rapid so that they can be successfully applied to any possible debris matrix and executed within the stringent timelines of a nuclear forensic response."

Seeing the Forest and the Trees

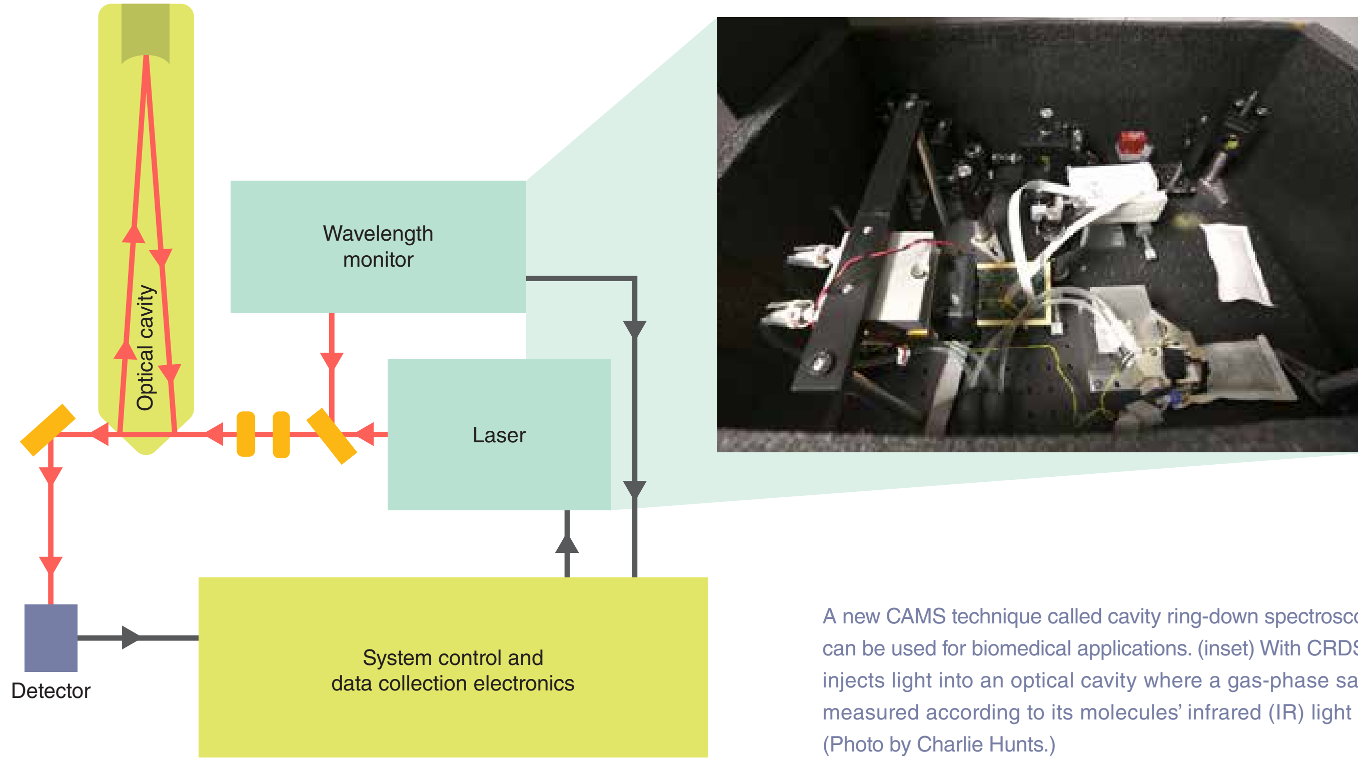
CAMS scientists conduct extensive radiocarbon analysis for multiple environmental science applications. Since joining the center in 2007, Earth scientist Karis McFarlane has studied carbon cycling in terrestrial ecosystems such as forests, wetlands, grasslands, and tundra. She works closely with DOE's Biological and Environmental Research (BER) Program on experiments that inform Earth system models. "We use radiocarbon analysis to track carbon as it moves through plants, soil, and the atmosphere," says McFarlane.

Through BER and the Laboratory Directed Research and Development Program, McFarlane and colleagues participate in a project called Spruce and Peatland Responses Under Changing Environments (SPRUCE). The multi-institutional study leverages CAMS's radiocarbon capabilities to measure ^{14}C and stable isotopes in carbon dioxide (CO_2) and methane emissions from a Minnesota peat bog. Scientists pump CO_2 and heat into enclosed sections of the peatland to simulate Earth's warming climate, testing the rates of carbon uptake, peat decomposition, and carbon release. "We see evidence that elevated temperatures increase both carbon uptake by vegetation and emissions, but in the short term we do not see losses of old peat carbon with warming," explains McFarlane. The rapid carbon cycling demonstrated by SPRUCE experiments help advance DOE's ability to predict and respond to climate change.

McFarlane's latest BER project—thanks to a DOE Early Career Research Program award—sends her to southern latitudes to better understand the role of moisture in carbon storage. For five years, her team will contribute to DOE's Energy Exascale Earth System Model (E3SM), a project that investigates complex processes such as the global water cycle and greenhouse gas fluctuations. (See *S&TR*, December 2017, pp. 4–11.) Soil samples from tropical forests in Puerto Rico, Cameroon, Indonesia, and several Latin American countries are being analyzed at CAMS for this project. "The tropics are chronically understudied along with ecosystem processes occurring below ground," notes McFarlane. "Tree root systems, for example, are not represented well in models. As a result, the models may have vertical inputs for roots going down only 20 centimeters, missing important soil carbon input patterns below that depth. Our soil profiles can help develop E3SM."

The Past Is Prologue

In addition to carbon-based studies, CAMS instruments are uniquely adapted for analysis of cosmogenic nuclides—the rare isotopes created in the atmosphere and terrestrial materials from cosmic ray collisions. With AMS, scientists can measure traces of these nuclides in rock, sand, sediment, and other natural



A new CAMS technique called cavity ring-down spectroscopy (CRDS) can be used for biomedical applications. (inset) With CRDS, a laser injects light into an optical cavity where a gas-phase sample is measured according to its molecules' infrared (IR) light absorption. (Photo by Charlie Hunts.)

materials to investigate the timing of processes such as glacial movement, landscape formation, tectonic motion, and climate change. For staff scientist Susan Zimmerman, an 11-year CAMS veteran, understanding these Earth system processes requires reconstructing the past. She says, "The goal is to bring the finest possible time resolution to the largest swath of Earth's history. This work is like flipping backward through a book to determine when events occurred and how fast changes have happened in the past."

"Fast" is a relative term to a geochronologist, and different cosmogenic nuclides provide specialized information about a specimen's historical surface exposure. For instance, ^{10}Be has a half-life of 1.4 million years, which makes it useful for analyzing old rock. Conversely, the half-life of beryllium-7 (^{7}Be) lasts only 53 days, leaving scientists with a fleeting measurement window before the isotope completely decays. Evaluating multiple isotopic signatures across a variety of sample materials helps Zimmerman piece together a more accurate picture of Earth's response to historical climate change. In one recent study, she was part of a team that measured aluminum and beryllium isotopes to chart erosion of the Greenland ice sheet as far back as 7.5 million years.

CAMS houses one of only two accelerators in the United States that routinely measures certain cosmogenic nuclides (^{26}Al, ^{10}Be, and ^{36}Cl), keeping staff busy running external users' samples as well as their own. Zimmerman, along with many CAMS scientists, often thinks about ways to expand the center's isotopic measurement capabilities to increase throughput as well as to extract more data from smaller sample sizes. She is working with staff scientist Alan Hidy and other Livermore colleagues to improve the efficiency of ^{7}Be measurement. Zimmerman says, "We can run a larger number of samples per day with greater sensitivity than before, so researchers can detect new patterns more quickly in the landscape they are studying."

Innovation with a Laser Focus

Since 1999, NIH has tasked CAMS with developing technology, analyzing samples, and interpreting data for the bioscience community. "Animal studies do not reveal everything about, for instance, how a drug will metabolize within a human. AMS's sensitivity allows us to quantify low-dose exposure," explains Turteltaub. "With consistent funding, we can move into the next phase as a user facility for bioAMS technology."

A laser-based system known as cavity ring-down spectroscopy (CRDS) is the newest tool in the bioAMS arsenal. CAMS staff scientist Daniel McCartt applies his engineering expertise to design and build better instrumentation. When McCartt was a Ph.D. student at Stanford University, he worked with CAMS and a commercial company, Picarro, Inc., to develop CRDS. He states, "Building a new instrument is not easy. At CAMS, I have access to analytical chemistry experts who can validate this new technology."

With CRDS, a laser injects light into an optical cavity where a gas-phase sample is measured according to its molecules' infrared (IR) light absorption. McCartt describes the cavity as a literal hall of mirrors. "When the laser is switched off, the light 'rings down' inside the cavity. We measure the light absorbed by the sample during this process," he explains. "The technique leverages the 'fingerprint region' of the mid-IR spectrum, where molecules are resonant with mid-IR light." The mirrors provide high sensitivity in a compact space, and the laser's specific wavelength required by the cavity length helps ensure high-fidelity measurements. Successful isotopic measurements include carbon-12, -13, and ^{14}C as well as the compound nitrous oxide.

The first-generation CRDS prototype garnered a patent and is undergoing testing. The system can record measurements every 2 to 15 minutes depending on isotopic concentration, and McCartt's team has achieved a run rate of more than 80 samples per day. McCartt is building a second-generation device to fit on a 0.60- by 1.82-meter tabletop. He states, "Our biggest challenge when trying to make a highly sensitive measurement is eliminating signal interference. The second CRDS version will use different spectroscopic techniques to better address these interferences."

The Next 30 Years

As CAMS embarks on its fourth decade of research, Bench predicts that AMS technology will become smaller, cheaper, and easier to use. He says, "We started biomedical work on tennis-court-sized instrumentation, and now we have a tabletop device." The center's efforts to make a resource more accessible over its lifespan mean that an AMS or CRDS device could one day be located where fieldwork is taking place or in a clinical setting. The bioAMS resource could be coupled with diagnostic imaging tools such as computed tomography and magnetic resonance imaging to provide a better understanding of disease trajectory and the effects of therapeutics on the body. Engineered human systems such as the Livermore-developed iCHIP (in vitro chip-based human investigational platform) present another valuable opportunity. (See *S&TR*, March 2014, pp. 16–19.) Turteltaub notes, "AMS has a role in addressing measurement challenges posed by these new technologies."

McFarlane looks forward to advances in radiocarbon extraction processes. "We are investigating simpler organic chemical extraction methods for natural abundance radiocarbon samples," she states. Tumey's group has begun developing additional nuclear forensic signatures. He says, "We want to structure sample-preparation protocols so that all isotopes relevant to AMS can be measured from a single, small aliquot of dissolved debris."

As technologies advance, CAMS is sure to retain its vibrant, innovative culture. Nanette Sorensen, the center's administrator since 1991, observes, "Something new is always happening. Our scientists like what they do." McFarlane confirms that research aimed at improving national security is "never boring," while Zimmerman underscores the interdisciplinary nature of the center's work. She explains, "We grow scientifically by working together, and this integrated view is exciting. We are all studying the same Earth."

—*Holly Auten*

A Reputation for Collaboration and Outreach

Over three decades, Lawrence Livermore's Center for Accelerator Mass Spectrometry (CAMS) has built a reputation for high-throughput analyses performed by staff with a wide knowledge base. A significant factor in the center's success is its emphasis on collaboration. "To be a credible user facility, it must be accessible and functional for external users," says founding director Jay Davis.

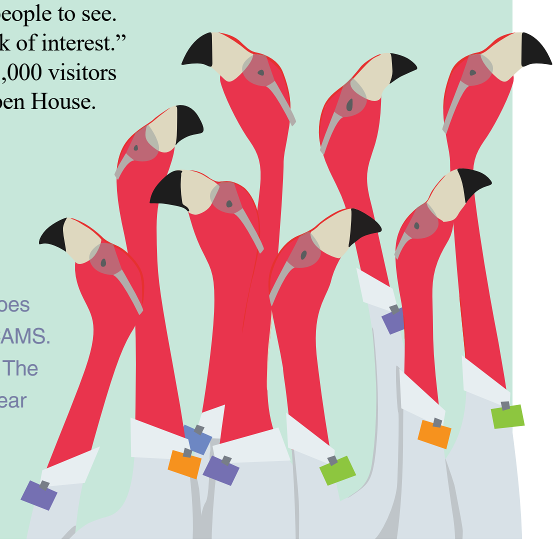
This open-door approach has helped CAMS secure multiple funding streams. The staff manage more than 200 work-for-others contracts and have participated in hundreds of national and international collaborations. Internal funding sources include projects underwritten by the Laboratory Directed Research and Development Program. "Lawrence Livermore is invested in codevelopment of novel technologies at CAMS because collaboration frequently leads to new ideas," explains CAMS director Graham Bench.

The center sees a steady influx of user-centered work from academic; commercial; and government partners, including other national laboratories. For some projects, external researchers send field-collected samples to CAMS for processing and data analysis. For other projects, scientists visit Livermore to observe their samples being processed with accelerator-based techniques, often relying on CAMS staff to help prepare specimens and interpret the data. In a third type of collaboration, CAMS scientists work with onsite Laboratory staff for specific projects, such as isotope production for the National Ignition Facility.

Initially conceived as the Multi-User Tandem Laboratory, the center was designed to be operated by scientists conducting research. According to Davis, many young researchers discover their potential through hands-on work at CAMS. He says, "This technology is sought after by the brightest students in the country." Ken Turteltaub, who arrived at the Laboratory in 1987 as a postdoctoral researcher and now leads the Laboratory's Biosciences and Biotechnology Division, notes, "At CAMS, scientists can work on scientifically risky problems and experiment with different technical approaches to solving them."

Over the years, CAMS has hosted approximately 1,300 faculty and students, becoming a gateway for potential employment at the Laboratory. Bench first worked with Livermore scientists as a graduate student in 1990. After seeing CAMS's capabilities up close, he asked for a job. More than two dozen Livermore careers began with similar stories at CAMS.

The center is also accessible to the public. In the past 2 years, staff have conducted more than 85 tours of the main accelerator facility for adults and regional schoolchildren. Nanette Sorensen, CAMS's administrator, states, "Our scientists volunteer as tour guides, which is exciting for young people to see. Engaging with a scientist could ignite a spark of interest." In October 2017, CAMS welcomed nearly 1,000 visitors during the Laboratory's 65th anniversary Open House.



The flamingo became CAMS's mascot after a series of practical jokes that began in the 1980s. From lawn ornaments and inflatable birds to plush toys and other trinkets, flamingoes inhabit the offices and laboratory spaces at CAMS. A bird even forms the "S" in the center's logo. The flamingoes shown here, wearing protective gear like their human counterparts, appeared on a flyer distributed at Lawrence Livermore's 65th anniversary Open House.

Prior to analysis, materials must undergo an extensive chemical pretreatment process to convert samples into a state of matter appropriate for the type of instrument used. Shown here, Livermore geologist Alan Hidy loads a sample into a centrifuge to separate its contents for future experiments. (Photo by Randy Wong.)

Key Words: acclerator, accelerator mass spectrometry (AMS), biological accelerator mass spectrometry (bioAMS), carbon cycle, cavity ring-down spectroscopy (CRDS), Center for Accelerator Mass Spectrometry (CAMS), climate change, cosmogenic nuclide, Energy Exascale Earth System Model (E3SM), environmental science, geochronology, infrared (IR) light, ion implantation, ion source, isotope, nuclear forensics, Nuclear Science User Facilities, paleoclimatology, radiocarbon, Spruce and Peatland Responses Under Changing Environments (SPRUCE).

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