*M*essage from the Assistant Deputy Administrator for Research, Development, Test, and Evaluation, Dr. Kathleen Alexander

The theme of this issue of the *Stockpile Stewardship Quarterly* is nuclear science. Specifically, we focus on nuclear data and the important role that this data plays in assessing the stockpile, in certifying warheads that have undergone a life extension program, and in nuclear forensics. This issue first highlights work being undertaken at Lawrence Livermore National Laboratory (LLNL) to iteratively improve the nuclear reaction data libraries through a formalized workflow process. These data libraries are essential for predicting nuclear reactions and decays. Not only is this data essential to understanding the performance of the U.S. nuclear stockpile, but it provides information applicable to any nuclear detonation, including that of an adversary.

One of the capabilities of the National Ignition Facility (NIF) at LLNL is to provide a neutron source to study neutron reactions (captures or fissions). Whereas neutron reactions can be measured at other facilities, such as particle accelerators, no other facility offers the neutron flux that the NIF produces; which is crucial for obtaining high-mass, lower yield products key to forensic detection of nuclear detonations. Additionally, NIF is tuned to produce a neutron spectrum peaked at thermonuclear neutron energy (14 MeV) which provides a unique platform to study high energy neutron reactions. Accurate measurements of fission-product yields obtained from the NIF platform can be used to assess the actinide fuel used in nuclear detonations.

We are pleased to announce four new Centers of Excellence awarded through the Stewardship Science Academic Alliance Program:

- Cornell University's Multi-University Center of Excellence for Pulsed-Power-Driven High Energy Density Science,
- Texas A&M University's Center for Excellence in Nuclear Training and University-based Research,
- University of Notre Dame's Actinide Center of Excellence (ACE), and
- the University of Texas at Austin Center for Astrophysical Plasma Properties.

Highlights about each Center of Excellence are provided. In addition, this issue includes a deep-dive article on ACE that highlights the unique and fascinating science taking place through this center to develop the skills and scientific understanding of actinide behavior crucial to the next-generation of stockpile stewards. Congratulations to the four new Centers of Excellence. We look forward to scientific advances from these Centers and to the pipeline of the next generation of stewards that this Center educates.

Last but not least, it is my great pleasure to announce that the inaugural director of the new Research, Development, Test, and Evaluation (RDT&E) Office of Experimental Sciences (NA-113) has been selected. RDT&E's Dr. Njema Frazier, who previously served as the Acting Director, was chosen from a pool of truly outstanding candidates. To learn more about Dr. Frazier and her impressive credentials, see page 8. The Office of Experimental Sciences, which includes the former Office of Inertial Confinement Fusion and Office of Research and Development, will allow us to offer even more value-added services to the Stockpile Stewardship Program. We have exciting plans for the remainder of the fiscal year. Thank you for exemplary work. Have a wonderful summer.

Dr. Njema Frazier

Inside

Better Nuclear Data Underpins Responsive Nuclear Enterprise by T.S. Bailey, B.R. Beck, C.M. Mattoon, and N.W. O'Brien (Lawrence Livermore National Laboratory)

If terrorists were to attack the United States with an improvised nuclear device, the ability of decision makers to convincingly determine who was behind the attack relies on a rapid and accurate assessment of the device by experts. Such an assessment depends on simulations that require high-quality nuclear data (see Figure 1).

Nuclear data is a common thread that underpins the safety and security of the U.S. nuclear stockpile, ensures safe operation of nuclear reactors, and is critical to reducing the threat of nuclear attack. Whereas researchers have worked for decades to measure fundamental data and to develop sophisticated nuclear theory, significant improvements still are required to improve the predictive capability of the data. Improved uncertainties in the data will improve the accuracy of assessments made by scientists when they are most urgently needed. Lawrence Livermore National Laboratory (LLNL) is revolutionizing its integrated nuclear data workflow to respond to a wide variety of needs as they are discovered.

Nuclear data are essential for predicting how nuclear reactions and decays occur to accurately model physical systems driven by neutrons and gamma rays. Examples of nuclear reactions range from scattering, where one particle "bounces" off another, to nuclear fission, where a collision between two particles causes one to split and release large amounts of energy. Each reaction can be described by the probability of that reaction occurring, known as a "cross section," and the outgoing energy and direction of each particle emitted by the reaction. Particles also may radioactively decay, emitting characteristic radiation that can be measured and used to identify specific materials.

To deliver high-quality nuclear data for National Nuclear Security Administration

Figure 1. Expert performs soil deposition monitoring using an Ortec Detective for the purpose of quantifying and identifying trace levels of radioactivity present to advise decision makers.
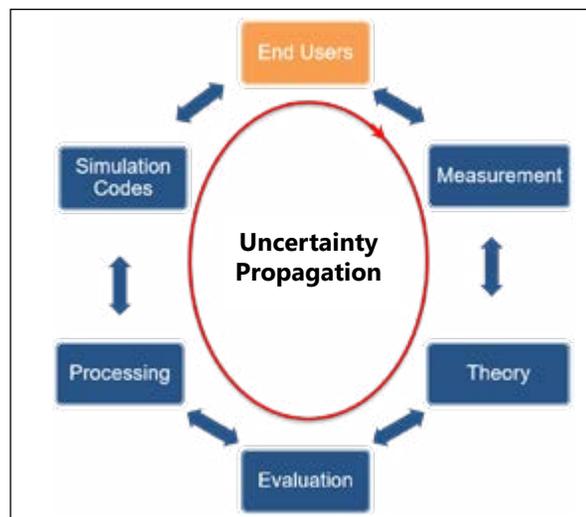

Figure 2. The nuclear data workflow is an iterative process of generating and improving the nuclear reaction data libraries that are used as inputs for end user applications.

(NNSA) programs, researchers at LLNL have developed a comprehensive, well-integrated nuclear data workflow. Key

features of the workflow, shown in Figure 2, include its iterative approach to generating nuclear data and specific quality control methods implemented to connect each step.

Components of the Workflow

The **end users** are the scientists and engineers who design and assess a wide variety of nuclear systems for NNSA programs. They are the customer of nuclear data, and they drive the nuclear data workflow by providing feedback and helping set priorities for their nuclear data needs. Nuclear data scientists rely on the end users to develop prioritized data needs.

Measurement involves designing and executing high-fidelity experiments to measure specific nuclear physics quantities of interest, including reaction cross sections and energy distributions for the particles produced during the reaction. New measurements often require significant investments in detection systems, target fabrication, and particle accelerators.

Nuclear theory supplements measurements by providing predictions for nuclear physics properties that are too difficult or expensive to measure and by interpolating and extrapolating beyond measurements to provide a complete description of each property. Theory is particularly important for developing data for short-lived, radioactive materials. The radioactive decay of these materials makes measurements extremely difficult because the target material disappears as it is being measured.

Evaluation is the process of combining experimental measurements and theoretical models into a set of recommended nuclear data with uncertainties. The U.S. evaluation effort is overseen by the U.S. Nuclear Data Program and involves contributions from multiple national labs, universities, and other
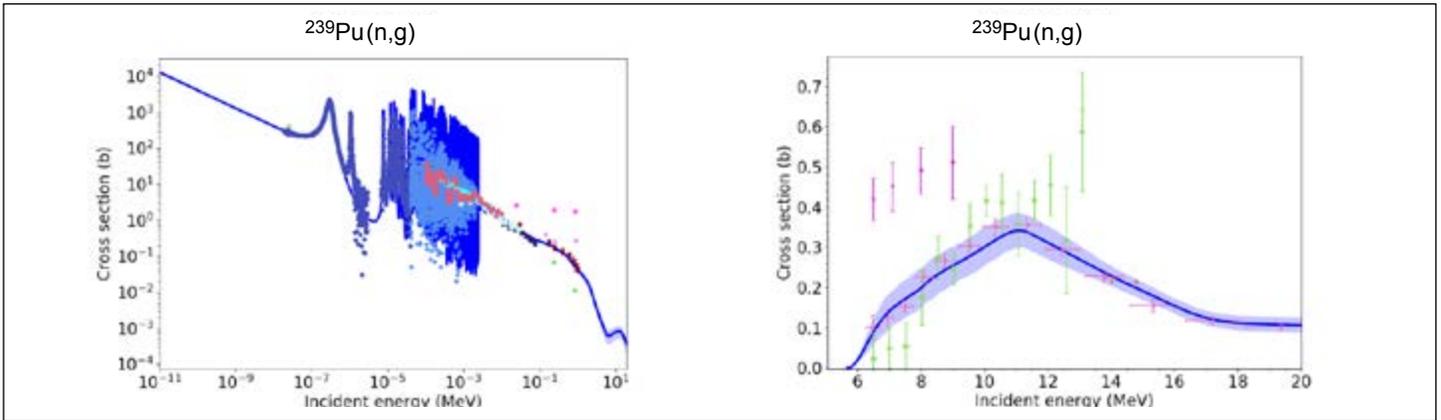
Figure 3. Two examples of reaction cross sections on plutonium-239 (^{239}Pu), showing the probability that each reaction will occur along with the uncertainty in that probability. Plots include experimental measurements (colored points) along with evaluated curves (blue lines) produced by combining nuclear theory with experiment.

institutions. The Cross Section Evaluators Working Group provides a forum for experimentalists, theorists, evaluators, processors, and users to collaborate on improving data libraries.

Processing transforms the evaluated data into a form suitable for use in particle transport codes. Even today the amount of nuclear data is far too large for the largest supercomputers. Processing steps include adjusting cross sections to account for material temperature, applying sophisticated averaging techniques to make simulation more tractable, and treating rapidly fluctuating cross sections (see Figure 3).

After processing, nuclear data are delivered to particle **transport codes**. These codes use the data to produce high-fidelity simulations of physical systems that are driven by neutrons and gamma rays. End users utilize these codes to model a wide variety of physical systems (see Figure 4).

Uncertainty propagation (UQ) ties the entire nuclear data workflow together, enabling end users to determine uncertainty estimates along with each prediction. UQ helps determine what nuclear data had the biggest impact in the simulation. This information is fed back to the rest of the pipeline, to identify the most significant nuclear data and to set priorities for new measurements, theoretical improvements, and evaluations.

The LLNL Nuclear Data and Theory (NDT) group has been modernizing the nuclear data workflow through improvements for

the evaluation, processing, and transport code steps. LLNL's NDT group has led the design of the new Generalized Nuclear Database Structure (GNDS), a replacement for decades-old nuclear data evaluation formats that were designed in the era of punch-cards. This LLNL-led GNDS design effort is an international collaboration involving experts from multiple U.S. and international institutions. The new structure stores both evaluated and processed nuclear data, making sharing data between institutions simpler. GNDS is easy to use and can be easily extended to handle new types of nuclear data that are identified as important by end users.

LLNL's processing and transport codes are in the final stages of being updated to accommodate the new GNDS data. When completed, this effort will enable higher-fidelity nuclear physics to be modeled with the transport codes. This effort also has led to improved testing and quality assurance across LLNL's nuclear data enterprise and a streamlined methodology for delivering new nuclear data to the end users at LLNL and other national laboratories. Testing includes comparing results using the latest (GNDS-enabled) processing and transport codes to earlier results using older formats and comparing evaluations before and after
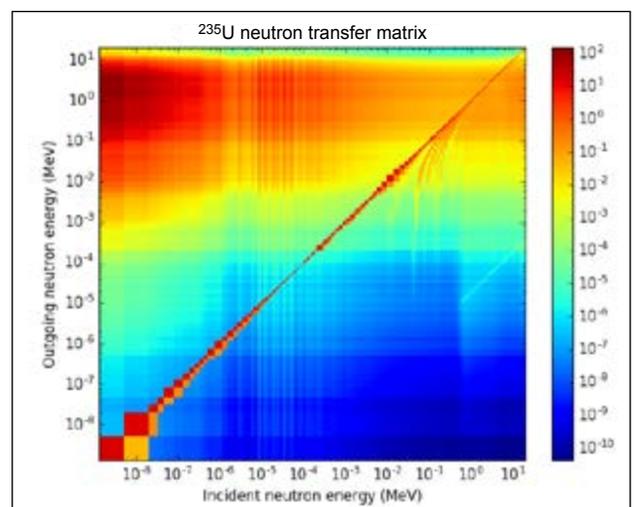

Figure 4. Nuclear reactions transfer energy from incident particles to outgoing reaction products. This figure shows a 'transfer matrix,' summarizing all possible reactions between a neutron and a uranium-235 (^{235}U) nucleus. The matrix can be used to efficiently model radiation transport in systems containing ^{235}U.

translating to ensure no data are lost during the translation.

As the stockpile ages and the nuclear threat environment evolves, the ability of researchers to be responsive is becoming increasingly important. Whether they are being asked to respond to a nuclear attack, assess the aging nuclear stockpile, or certify the safety, security, and effectiveness of warheads that have gone through life extension programs, the ability of researchers to meet the national security needs of tomorrow will rely on better access to improved nuclear data. LLNL and its NDT group are taking the lead in making sure the nuclear security enterprise is ready to answer the call. ●

Development of Platforms for Nuclear Science Experiments at the National Ignition Facility

by D.A. Shaughnessy, K.J. Moody, N. Gharibyan, J.D. Despotopulos, and P.M. Grant (Lawrence Livermore National Laboratory)

Introduction

When neutrons are produced in nuclear processes such as fission or fusion, they can interact with any surrounding materials, inducing radioactivity through neutron capture reactions. Neutron reactions are fundamental to many areas of nuclear science, including studies of nuclear reactors, determining the formation of matter in stars, nuclear forensics, medical radiotherapy, and assessing the safety of our nuclear stockpile. The National Ignition Facility (NIF) offers a unique opportunity to use the inertial confinement fusion platform to measure neutron reactions essential to national security or fundamental and applied science applications. A typical fusion experiment at NIF currently produces approximately 10^{16} neutrons through the fusion of deuterium and tritium in a target capsule. Materials introduced into the NIF target chamber in close proximity to the capsule will undergo neutron reactions such as capture or fission, and the resultant reaction rates can be evaluated.

Whereas neutron reactions can be measured at other facilities, NIF offers unique advantages. The flux of neutrons produced (number of neutrons per square centimeter per second) is much higher than any other available neutron source due to both the small size of the NIF capsule (2-mm diameter before implosion) and the short time scale (roughly 100 ps) over which the neutrons are produced. This results in less target material required to perform a reaction-rate study (less than or equal to 10^{16} atoms) as compared to traditional particle accelerators which require targets that are approximately 1 mg. In addition, the NIF neutron spectrum is peaked at thermonuclear (14 MeV) neutron energy. This means that nuclear reactions with nonthermal energy thresholds can be studied at NIF, whereas neutron energies at a nuclear reactor generally are insufficient for studying higher-energy neutron reactions. Several experimental configurations have been developed that enable the use of NIF fusion capsules as powerful neutron sources for nuclear reaction measurements.
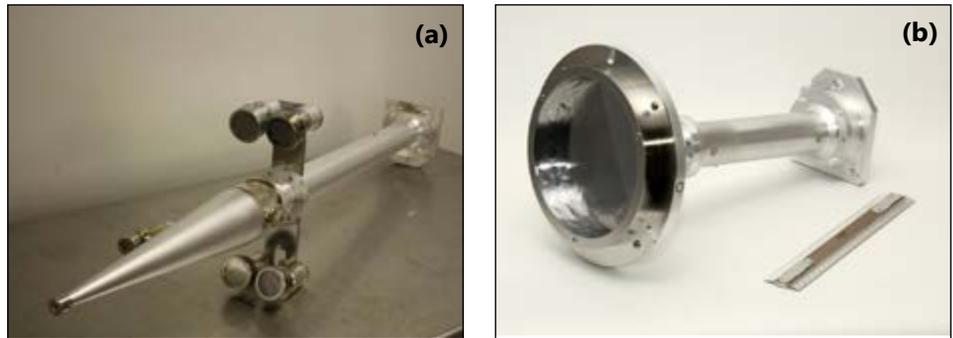
Figure 1. (a) Four SRC collector plates mounted on the end of a nose cone that is later attached to a Diagnostic Instrument Manipulator. In this configuration, the 50-mm-diameter metal discs are 50 cm from the NIF capsule. (b) The Large Area Solid Radiochemistry collector. When this configuration is fielded 50 cm from the NIF capsule, the collector surface covers roughly 1% of the 4π solid-angle. After a NIF shot, these collectors are retrieved within 2 hours for radiochemical processing and radiation counting of the collected debris.

Nuclear Science Platforms

To perform fundamental nuclear science measurements at NIF, materials first must be introduced into the chamber for irradiation, then retrieved post-shot to extract activated target debris, radiochemically processed to optimize sample content, and measured via radiation counting. The reaction products produced during a NIF shot are initially in a plasma environment and subsequently condense upon cooling into solid particulates that are collected on metal plates fielded inside the chamber 50 cm from the exploding capsule. The Solid Radiochemistry (SRC) diagnostic uses 50-mm-diameter collectors mounted on the end of a Diagnostic Instrument Manipulator.[1] There also is an option to field a larger diameter collector that covers an area roughly 1% of the 4π solid-angle (see Figure 1).[2] Through previous measurements, it was determined that collection efficiency of the post-shot debris is roughly proportional, or perhaps even slightly greater, than the solid-angle of the collecting plate. These collectors are retrieved approximately 2 hours after a NIF shot, when they either may be dissolved for radiochemical processing of specific reaction products or analyzed directly with high-purity germanium detectors through nondestructive gamma-ray spectroscopy.

Collectors are used when a NIF capsule has material placed in or around it for nuclear reactions to occur in close proximity to the source of neutrons, thereby taking advantage of the greatest neutron flux. For some measurements, it is sufficient to have encapsulated targets located some distance from the neutron source during irradiation. The Target Option Activation Device (TOAD) is a sealed container that is used to irradiate materials positioned 50 cm from the center of the NIF chamber. The TOAD may be fielded in different configurations to accommodate diverse types of materials and experimental protocols (see Figure 2).

Nuclear Reaction Measurements

Reactions that have been studied to date include neutron-induced fission and activation reactions where one neutron is absorbed and two are subsequently emitted (the (n,2n) reaction). When an actinide nucleus undergoes fission, it becomes two lighter-mass fission fragments. The identity of these fragments is determined by their fission product yields, which is the fraction of fission events that results in a particular isotopic species (because each fission produces two primary fragments, the yields total to 200%). Accurate measurements of these fission-product yields are essential for post-detonation weapons diagnostics, which use these data to convert the measured fission

products in post-explosion debris into an assessment of the actinide fuel that was used in the device. The short pulse of high-flux neutrons that occurs in a NIF shot can irradiate actinide materials in a TOAD target holder and measure fission yields with better fidelity and accuracy.

As an example, samples of uranium-238 (^{238}U) were placed in TOAD holders 50 cm from the NIF target chamber center. After irradiation, samples were recovered, and the fission products were measured by a combination of radiochemical separations and radiation counting. Figure 3 shows the fission-product yields that were measured using this technique.[3] Of note are the number of low-yield products

at the highest mass numbers that could be determined due to the NIF neutron flux. With less intense neutron sources, these "wing" products are not accessible by irradiation measurements, but their fission-product yield data are imperative for post-detonation nuclear forensics because of their extreme sensitivity to the identity of the fissioning fuel.

Accurate measurements of (n,2n) reactions are important for validating models used to determine device performance based on previous nuclear testing data. Reaction cross sections typically are available for first-order reactions on stable nuclides, but in a prompt, high-flux environment such as

a nuclear test, second-order reactions also can occur when excited nuclear states formed by the first-order reaction undergo subsequent (n,2n) reactions. One of the few ways to measure such cross sections of short-lived excited nuclear states is to add material to a NIF capsule in close proximity to the fusion neutrons and measure the resultant reaction products. Currently, LLNL is developing methods to inject up to 10^{16} atoms of a radioactive species on the inner surface of a NIF capsule. The first capsules tested with this method will have ^{238}U deposited on the inner surface to compare to results obtained from TOAD measurements as described above. Once the method has been validated, yttrium will be used for the first (n,2n) measurements on a shorter-lived radionuclide. Two capsules will be prepared—one with stable yttrium-89 deposited on the inner surface and one similarly prepared with radioactive yttrium-88 (^{88}Y). The differences in the measured reaction cross sections between the two will be attributed to the contributions from excited nuclear states in ^{88}Y, which currently are unknown. Additional isotopes that were used as thermonuclear performance indicators during nuclear testing also will be examined at NIF.
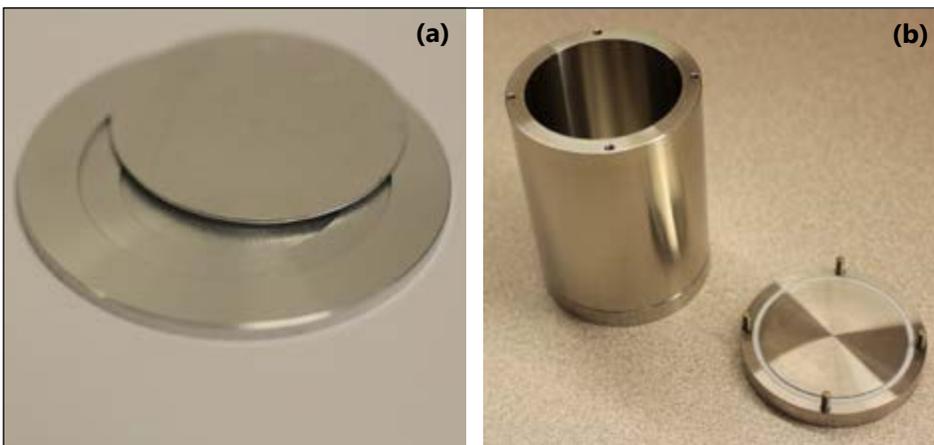

Figure 2. (a) The TOAD is used to field foils for neutron irradiation 50 cm from the NIF capsule. Foils are sealed inside the TOAD, which is removed after a shot for processing. (b) The H-TOAD container is used to similarly field several grams of material or multiple foil stacks.
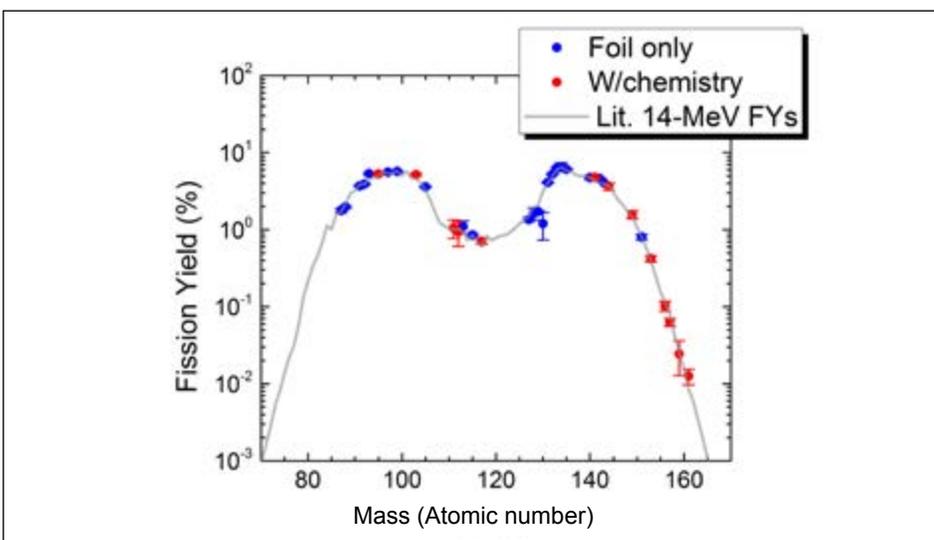
Figure 3. Results from TOAD irradiations of ^{238}U. After neutron irradiation, the ^{238}U foils were radiochemically processed (red dots) or assayed for gamma radiation (blue dots). The line is the evaluated fission yield data for ^{238}U.[4] The high-mass nuclides are produced with lower yields and can be observed only in high-flux environments.

Conclusions

Through development of radiochemical diagnostics and microinjection of target materials into NIF capsules, experimental platforms have been developed that can be used to measure fundamental nuclear reactions at NIF. Future measurements will focus on (n,2n) reactions on excited nuclear states, which only can be measured in high-flux environments, such as a NIF capsule.

References

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[2]C. Waltz et al., Target Diagnostics Physics and Engineering for Inertial Confinement Fusion VI, J.A. Koch and G.P. Grim, eds., Proc. of SPIE 10390, 103900H (2017).

[3]N. Gharibyan et al., J. Radioanal. Nucl. Chem. 303, 1335 (2015).

[4]T.R. England and B.F. Rider, Evaluation and Compilation of Fission Product Yields 1993. Technical Report LA-UR-3106, Los Alamos National Laboratory (1995). ●

Compensating Errors in Nuclear Data *by Morgan White (Los Alamos National Laboratory)*

Just over a decade ago, the weapons science community came together to face the challenge of quantifying uncertainties in predictions of nuclear weapons performance resulting from the underlying data in our simulations. As the Advanced Simulation and Computing initiative continued to deliver ever larger and faster computers and the new codes to take advantage of them, it became time to evaluate the answers. This work showed such simulations were sensitive to the data, and there were many questions where nuclear data uncertainties were significant. This is not surprising to those who use simulations daily. Computers always should display the warning label "garbage in, garbage out." These studies motivated the efforts that have since become known as the Advanced Fission Measurements campaign.

Neutron reactivity is central to the fields of criticality safety, nuclear reactors, and nuclear weapons. These fields correspond to the subcritical, critical, and supercritical reactivity states, respectively, and apply during the manufacture, production, and use of special nuclear material. A critical system is one in which the number of fissions is constant. This condition corresponds to the steady-state nature of an operating nuclear reactor. The safety of personnel in facilities that handle nuclear materials requires keeping nuclear materials from reaching a critical state in order to prevent radiation accidents. Nuclear weapons seek to make militarily significant quantities of energy through an exponential cascade of fissions. Our ability to predict these conditions requires knowledge of the underlying physics.

Nuclear reactors only are possible because of delayed neutrons from fission. After a nucleus fission, neutrons are emitted on prompt (picosecond or faster) and delayed (millisecond and longer) time scales. Steady-state nuclear reactor operations occur when the number of fissions from all neutron interactions are constant. This is called delayed critical. Operating

slightly above delayed critical, the number of fissions increases on time scales corresponding to the delayed neutron emissions and, therefore, is controllable through mechanical interactions. Severe criticality safety accidents and nuclear weapons operate in states above prompt critical, where prompt critical represents the theoretical state of a constant fission rate only from prompt neutrons. Not long before he lost his life to a prompt criticality accident, Louis Slotin coined the term *one dollar* as the unit interval
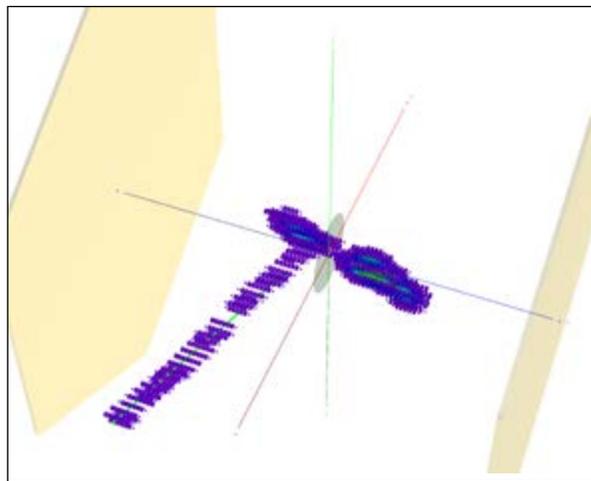

Figure 1. The fission TPC brings techniques pioneered by high-energy physics to study the fundamental particles of nature to provide a camera-like view of the fission process. This track reconstruction shows a ternary (two fission fragments and an alpha particle) fission event.

between delayed and prompt critical. Reactor physicists still use dollars and cents to discuss neutron reactivity.

Our original goal motivated by the earlier uncertainty studies was to better predict neutron reactivity. There are several reactions that dominate this process: how often a fission occurs (defined by the fission cross section), the number of neutrons emitted (aka nubar), their spectrum of energies (aka prompt fission neutron spectrum [PFNS]), the likelihood they are absorbed (the capture cross section), and their likelihood of escape (influenced by the scattering cross sections). As you might expect, given their obvious importance, the key fission cross sections have been measured many times over many decades. The consensus among experimentalists who measure these data

judged that the best of the fission cross section measurements had an accuracy of approximately 1-2% (which corresponds to dollars in reactivity uncertainty). But they also warned that many potential experimental errors easily could exceed this estimate. Despite these misgivings, standard practice used the least squares method to fit these data and produced residual error estimates of 1% or less. There was considerable skepticism about using such low derived uncertainty estimates.

Scientists at Lawrence Livermore National Laboratory (LLNL), Los Alamos National Laboratory (LANL), and several other laboratories and universities had a plan to bring new techniques to tackle measurement of the fission cross section. LLNL designed fission Time Projection Chamber (TPC) use techniques pioneered by high-energy physicists in pursuit of the building blocks of matter to shed new light on the fission of the atom (see Figure 1). Historic detectors used simple methods to monitor the large energy pulse induced by the two fission fragments. Many measurement errors involve a failure to correctly discriminate fission fragments that have lost much of their energy from other energetic particles (e.g., alpha decays or scattered particles). Other measurement errors involve quantifying the efficiency of detecting these events. The fission TPC provides detailed images of these events that allow us to study the causes of many of these errors. The fission TPC project has shed remarkable insights on both the new measurements and previously misunderstood issues with historic measurements.

Despite the potential uncertainties in our nuclear weapons analyses, we always have been able to match simulation results to many experimental measurements across a wide range of tests. As we considered that the fission cross section might be wrong, we recognized that a change in its values would require uncovering some other unknown issue. Otherwise, the changes would cause our suite of good simulations to produce bad results. We
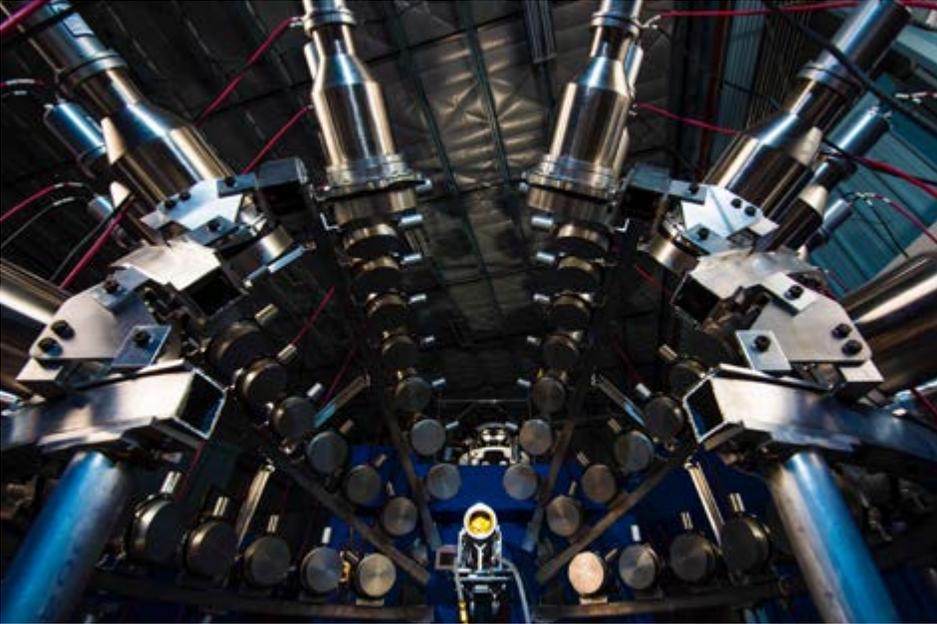
Figure 2. The Chi-Nu detector array. The Los Alamos Neutron Science Center facility produces 'white spectrum' neutrons that interact with a fissionable target (golden disc near center) and are subsequently detected by an array of liquid scintillator (shown) or lithium-glass detectors. The double time-of-flight measurement technique uses the times from the source to target to detection to infer the PFNS as a function of the incident neutron energy.

Figure 3. The Lady Godiva (high-enriched uranium) critical assembly experiment after a prompt supercritical accident involving 5 cents of excess reactivity. The National Criticality Safety Program operates the National Critical Experiments Research Center in Nevada.

now refer generally to this issue by the term compensating errors. It is not that our simulations are truly predictive, rather we have numerous compensating errors that offset each other. That is, given that we can match enough data, we believe similar simulations predict many aspects of events that are near neighbors. However, if we change one part of the data in the simulation, even if we know the change is for the better, this balance is upset and the system is less predictive. So any change to the fundamental data used in the analyses must be accompanied by at least one other change to maintain the calibrated predictive capability.

Understanding that we could not make a single change, we started a second major measurement effort. The Chi-Nu project, a joint LANL and LLNL undertaking, expanded a previous effort to measure the prompt fission neutron spectra (which had been shown to have uncertainties similar to the fission cross section) (see Figure 2). As new techniques were not available, the goal of this effort was not just to make new measurements of missing data, but to systematically study the errors involved in both the current and historic measurements. These efforts resulted in both new measured data and, perhaps even more importantly, insights that unraveled many discrepancies

between historical data sets. This change in perspective, from seeking to measure a mean value to trying to quantify our errors, has been slowly making its way into all aspects of our ongoing measurement program.

Every few years, it is important to put down a marker that defines where you are, if for no other reason than to be able to trace where you have been. In the United States, the National Nuclear Data Center Cross Section Evaluation Working Group produces the ENDF/B nuclear data library. The eighth major version, ENDF/B-VIII.0, was released in February 2018. In addition to its contributions to the ENDF/B library, the NNSA Defense Programs nuclear data projects also made major contributions to new international nuclear data standards coordinated by the International Atomic Energy Agency. A special edition of the journal *Nuclear Data Sheets*, Volume 148 (March 2018), documents this work and includes further references to dozens of key contributions over the last decade. Among this substantial body of work can be seen an emerging consensus regarding the uncertainties within these data and their impact on applications.

Perhaps most shocking to those outside the community will be that estimates of

uncertainties on many of the best known nuclear data quantities have not been reduced; rather they are now double, quadruple, or even higher. When we use mathematics to rigorously define our certainty, we must be careful to appreciate its limitations. Richard Feynman is famous for his argument that it does not matter how beautiful and elegant your theory is. If it does not match experiment, it's wrong. Our first reaction to this critique is to continue to develop the theory until it is correct, as proven by how well it matches the experiment. We now realize that this simplified response does not go far enough. Compensating errors mean that there are many ways to be correct. For example, predicting a 5-cent reactivity swing involves seven major nuclear reaction components, three of which have uncertainties of the order 10 cents to a dollar and four of which have uncertainties of one or more dollars (see Figure 3). It is not good enough to prove the accuracy of the mean using a single choice of these values. What we are truly after is to prove the negative. To do so, we would have to examine all of the data combinations used to predict a reactivity state in order to prove that none of them led to an unexpected failure.

It is likely impossible, at least in the near future, that we can measure the fundamental nuclear data needed for

many analyses to the level that they are accurate enough on their own. But this need is not to say that our job is hopeless. Over the last hundred years, the advances in nuclear physics have built and maintained a nuclear weapons deterrent, built nuclear reactors for both power and propulsion, developed nuclear medicine to a maturity that it is a major diagnostic and therapeutic tool, and provided cosmology the tools to illuminate the origins of matter in the universe. These accomplishments did not require us to be perfect. Rather they required, and still require, us to better comprehend these limitations and find ways to go beyond them. In practice, we build critical assembly experiments to calibrate computational models to near neighbors of our end applications. This calibration

has always been implicit within our work. Our recent understanding amounts to rediscovering our reliance on this process. Many fields of engineering face similar challenges and have developed methods we need to adopt to allow us to quantify the limitations of extrapolations around these calibrations.

The increased uncertainties associated with our fundamental data reinforce long-standing observations that there are many computational models that obtain equivalent results for a given analysis. While necessary, measurements of the fundamental data on their own will not achieve an accuracy to discriminate between these models. To shed more light on and expand the elusive boundary between good and bad predictions, we

must find novel new differential and integral experiments—or make better use of our historical measurements—to further constrain the relationships between the components in these models. The Advanced Fission Measurement campaign has provided a deeper appreciation of the role of nuclear data within this process and pointed the way towards many new lines of study. NNSA Defense Programs and its Stewardship Science Academic Programs university partners continue to deliver a diverse suite of nuclear science research that will strengthen our ability to perform our stockpile stewardship and global security missions, as well as support the broader nuclear science community. ●

Meet the Director, Office of Experimental Sciences: Dr. Njema Frazier

We are pleased to welcome Dr. Njema Frazier as a member of the Senior Executive Service, serving as the Director of the Office of Experimental Sciences (NA-113). As Director of NA-113, she will oversee the Inertial Confinement Fusion portfolio and a large fraction of the Science portfolio.

Dr. Njema Frazier is a theoretical nuclear physicist in the NNSA's Office of Research, Development, Test and Evaluation. She has been a member of Defense Programs since 2001 and has served as Physicist, Acting Deputy, and Acting Director for a number of NNSA's flagship scientific and technical programs established to ensure the United States maintains a safe, secure, and effective nuclear weapons stockpile without explosive testing. These include positions in the Office of Advanced Simulation and Computing (program manager and acting Deputy), the Office of Defense Sciences (program manager for Secondary Assessments as well as acting Director for International Programs), and Office of Inertial Confinement Fusion (acting Director).

In her 20-plus years of federal service, Dr. Frazier has been the recipient of multiple career and national awards, including the Department of Defense Joint Civilian Service Commendation Award;

the Award for Distinguished Service to the National Nuclear Security Administration (NNSA); the Black Engineer of the Year award, Science Spectrum's Trailblazer Award; the EBONY Power 100 list, Ebony Magazine's annual list of the nation's most influential African Americans (Frazier pictured above on the Red Carpet for the event); and the Black Girls Rock! Awards where she was honored as the STEM Tech Recipient for 2017. She also was recently honored by Carnegie Mellon University with an Alumni Achievement Award.

Prior to joining the NNSA, Frazier was a professional staff member for the U.S. House of Representatives Committee on Science for four years. While at NNSA, she was a Visiting Professor at the National Defense University, College of International Security Affairs for three years. She was the first African-American woman to graduate with a physics degree

Dr. Frazier was one of the "Nifty Fifty" innovators and role models in science, technology, engineering, and math (STEM) at the 2014 USA Science & Engineering Festival. The program provides middle and high school students in the Washington, DC region with interactive, hands-on STEM experiences.

from Carnegie Mellon University, as well as the first to receive a PhD in nuclear physics from Michigan State University. She was the co-founder of the Department of Energy POWER (Professional Opportunities for Women at Energy Realized) Employee Resource Group, a member of the National Advisory Board of the National Society of Black Engineers, and chair of the Algebra by 7th Grade Initiative for grades 3 to 7.

Please join us in welcoming Dr. Frazier to her new position in Defense Programs. ●

University of Notre Dame Actinide Center of Excellence *by Peter C. Burns (University of Notre Dame)*

The newly established Actinide Center of Excellence (ACE), led by the University of Notre Dame and directed by Peter C. Burns, focuses on graduate students and postdoctoral researchers working together with distinguished professors developing a fundamental scientific understanding of the chemistry of the actinide elements. Actinides occupy the bottom row of the periodic table in which all elements are arranged according to the filling of electron orbitals. The actinides correspond to the sequential filling of the 5f electron orbitals, and they include the heaviest natural elements. All of them are radioactive, but the decay rates of thorium and uranium are long enough that they persist from the formation of Earth about 4.5 billion years ago. Protactinium, which is between thorium and uranium in the periodic table, results from radioactive decay of uranium and is natural. All of the actinides heavier than uranium (transuranium elements) are synthesized in a cyclotron or nuclear reactor, and the first two, neptunium and plutonium, were discovered in 1940 and named after planets (as was uranium 150 years prior).

Understanding the chemistry of actinides is essential for our Nation because they are the fuels of nuclear weapons and also of reactors that generate 20% of our electricity, propel many naval vessels, and produce medical isotopes. They are environmental contaminants where uranium or thorium was mined or processed and where plutonium was produced for weapons. For Stockpile Stewardship, understanding the fundamentals of actinide chemistry helps to predict aging of weapons components that impact functionality and is essential for the processing and manipulation of these elements in all of their forms.

Chemistry is a discipline that focuses on the compositions, properties, and reactivity of matter. Many chemists work towards understanding and controlling chemical reactions in which elements bond to each other to produce materials with useful properties. The emphasis is on the chemical bonds between elements, and the elements themselves do not

change. Radiochemists similarly deal with chemical bonding, but because the elements are radioactive, they change their identities over time to other elements, and they release several forms of potentially hazardous radiation. Some isotopes of lighter actinides decay slowly, and their gradually changing identity is not an issue for doing chemical experiments that are on a shorter time-scale. However, all actinides are radioactive, and the radiation emanating from a sample during an experiment is potentially hazardous. The radiation often causes changes in the chemistry of the system. For example, the deposition of energy in water from ionizing radiation produces several reactive chemical species, including hydrogen peroxide, that have very different properties than water.
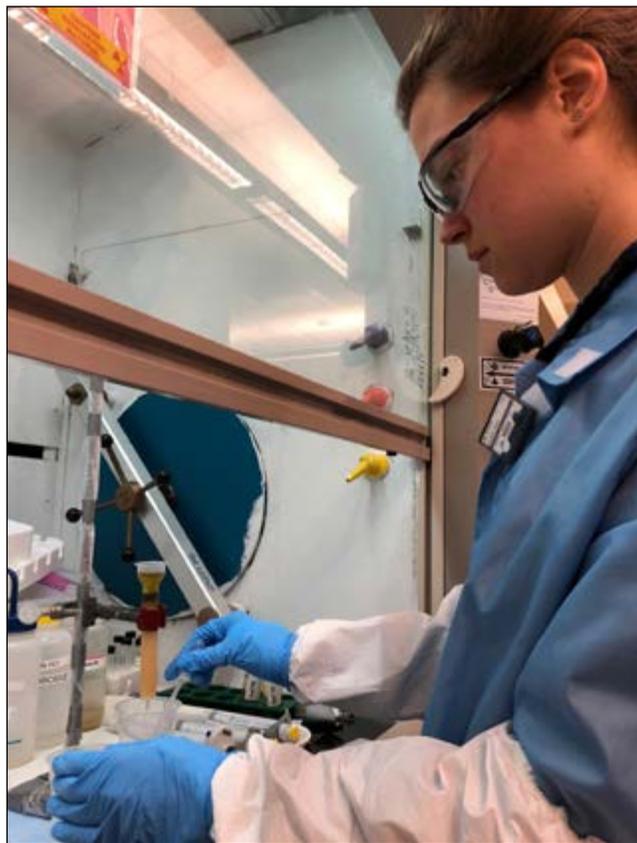

Figure 1. Sara Gilson, PhD student in chemistry and biochemistry at the University of Notre Dame, loads a neptunium-bearing solution (green) into a column for purification.

Doing chemistry with actinides is difficult, because they are extremely complex and have many different oxidation states and chemical forms. Scientists must be protected from the radiation they emit. Unlike uranium and thorium that are readily available from geologic deposits, the transuranium elements must be synthesized at great expense and are strategically very important, which strictly limits their availability. Doing research with actinides generally is much harder and more expensive than elsewhere on the periodic table and requires dedicated facilities and highly-trained personnel. Most university chemistry departments throughout the world have no program in actinide chemistry, and even undergraduate courses that emphasize actinides or radiochemistry are rare. Yet there is

a great need for actinide expertise in Stockpile Stewardship, and this is being addressed by the newly founded ACE.

ACE will consist of 16 PhD graduate students, 8 postdoctoral scholars, and 8 professors at the University of Notre Dame, Oregon State University, Northwestern University, Washington State University, and the University of Minnesota. Students gain hands-on experience planning and executing experiments and computations in actinide chemistry, working safely with radioactive materials, and are trained in the use of a broad range of materials and chemical characterization methods (see Figure 1). Every student in the program will conduct part of their research in an National Nuclear Security Administration national laboratory, and ACE provides abundant opportunities for students to network
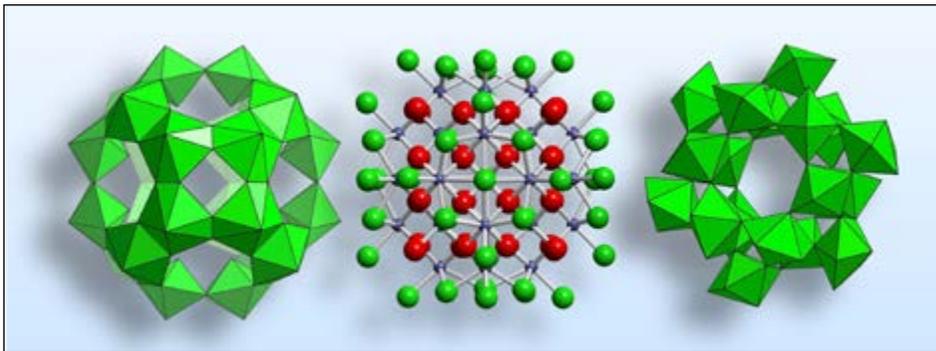
Figure 2. Examples of actinide oxide clusters discovered by ACE researchers. From left to right: A cage containing 24 neptunium atoms, a nanocluster with a core consisting of 38 plutonium atoms and 56 oxygen atoms, and a crown containing 18 neptunium atoms.

Figure 3. A large calorimeter at the University of Notre Dame is dedicated to studies of actinide chemistry and is the only such instrument in the world that can be used for measurements of transuranium compounds.

with leading scientists nationally and internationally. Six distinguished scientists from other universities and national laboratories serve on the Scientific Advisory Committee, providing oversight of ACE activities.

Much of the research emphasis of ACE is focused on actinide-based nanoscale clusters (see Figure 2). Made up of tens to hundreds of atoms and with sizes about 1/100,000 the width of a human hair, actinide nanoclusters are bigger than single atoms and have special chemical properties that are often dependent on size, shape, and structure. Researchers in ACE are pioneers in the study of actinide oxide clusters and have discovered approximately 100 unique cluster types. They synthesize actinide oxide clusters and study their chemical properties, sizes, and atomic-scale structures. Clusters studied range in composition from uranium and neptunium peroxide hydroxides through plutonium oxide chloride clusters that have well-defined structures and properties that are typically dramatically different from those of simple actinide cations in solution. These clusters are powerful models for studying actinide chemistry, because properties can be enhanced at the nanoscale.

It is becoming increasingly more feasible to harness the complex properties of actinide oxide nanoclusters for specific purposes. Applications of actinide oxide nanoclusters under study by ACE researchers include: 1) separations of chemical constituents of complex radioactive materials, 2) dissolution of normally highly insoluble actinide materials, and 3) the production of porous materials for capturing and filtering at the nanoscale. Because many of the potential applications of these materials are in highly radioactive environments, ongoing work is determining the effects of different types of radiation on these materials.

Researchers in ACE study nanoscale actinide clusters using x-ray, laser, and neutron beams in order to understand their formation mechanisms, properties, and structures. Experiments are complemented and inspired by quantum mechanical calculations and molecular dynamics simulations, which allow prediction of the chemical behavior, energetics, and structures in these systems. ACE researchers actively are improving these computational methods and are developing data for real systems to benchmark computations.

ACE researchers measure the energetics of different transformations that occur in actinide chemistry so that the predictive powers of thermodynamic laws can be applied. A one-of-a-kind facility at the University of Notre Dame has five calorimeters for measuring the energetics of actinide systems. Two room-filling calorimeters operate at 700 °C and are so sensitive that they can perform measurements for samples that are only 0.005 grams (see Figure 3). Techniques for working with very small samples are essential in actinide radiochemistry because they reduce the quantity of radiation that must be managed during experiments. ACE has several spectrometers mounted on microscopes and x-ray instruments with intense beams for studying very small samples.

By focusing on the chemistry of actinides at the nanoscale, ACE students gain experience in a wide range of advanced computational and experiment techniques and utilize national user facilities, including synchrotrons and neutron sources. They develop a unique set of skills and scientific understanding of the behavior of actinides that prepares them for scientific careers related to national security or elsewhere. ●

Suggestions or Comments?

Do you have a great idea for an article or comments about this issue of the *Stockpile Stewardship Quarterly*? If so, please email Terri Stone at terri.stone@nnsa.doe.gov.

NNSA Names Four New Centers of Excellence

The Department of Energy's National Nuclear Security Administration (NNSA) has named four new Centers of Excellence within the Stewardship Science Academic Alliances (SSAA) Program: Cornell University, Texas A&M University, the University of Notre Dame, and the University of Texas at Austin. An NNSA Center of Excellence is a multi-investigator team, made up of multiple academic institutions, to address an over-arching theme or themes of interest within a topical research area relevant to the Stockpile Stewardship Program. Centers of Excellence attract high caliber graduate students and train them in areas critical to stockpile stewardship, providing students with exposure to the research community and the NNSA national laboratories. The new Centers of Excellence are discussed in the following paragraphs.

Cornell University's Multi-University Center of Excellence for Pulsed-Power-Driven High Energy Density Science will be led by Drs. David Hammer and Bruce Kusse. The mission of this Center is to carry out fundamental studies of High Energy Density (HED) plasmas produced by pulsed power generators. The principal objectives of this multi-university research center are to improve the understanding of the properties of dense, high temperature plasmas, especially in the presence of strong magnetic fields, while training the next generation of HED research scientists. Partner institutions are Imperial College, Weizmann Institute of Science, University of Michigan, Princeton University, University of California, San Diego, Lebedev Physical Institute, and the University of New Mexico.

The Center for Excellence in Nuclear Training and University-based Research (CENTAUR), led by Dr. Sherry Yennello of Texas A&M University (TAMU), will pursue investigations in low-energy nuclear science, including experimental, theoretical, and technical programs using facilities at the Cyclotron Institute at TAMU and the John D. Fox Accelerator Laboratory at Florida State University. Existing collaborations between scientists at TAMU and the NNSA national
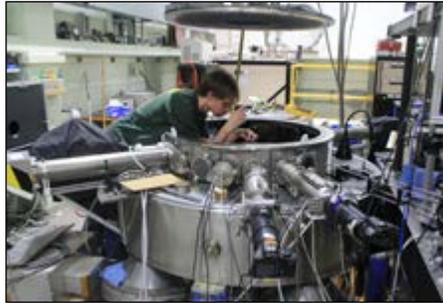

Cornell University PhD candidate Jacob Banasek preparing a load for Cornell's one million ampere generator, COBRA machine.

TAMU student Layla Bakhtiari preparing equipment for an experiment.

laboratories will be incorporated into the center program and expanded to involve scientists from the partner institutions: Florida State University, Washington University, University of Washington, and Louisiana State University.

Research conducted at the Actinide Center of Excellence (ACE), led by Dr. Peter Burns of the University of Notre Dame, will integrate both experimental and computational approaches to investigate radioactive materials, including americium, neptunium, plutonium, and uranium, taking advantage of specialized facilities developed at Notre Dame. Further, the team of researchers will focus on three specific themes: the properties and structure of actinide-oxide clusters, the thermochemistry of actinide materials, and how nanoscale nuclear materials react in various chemical environments. They will collaborate with other renowned research institutions, including Northwestern University, Oregon State University, the University of Minnesota,
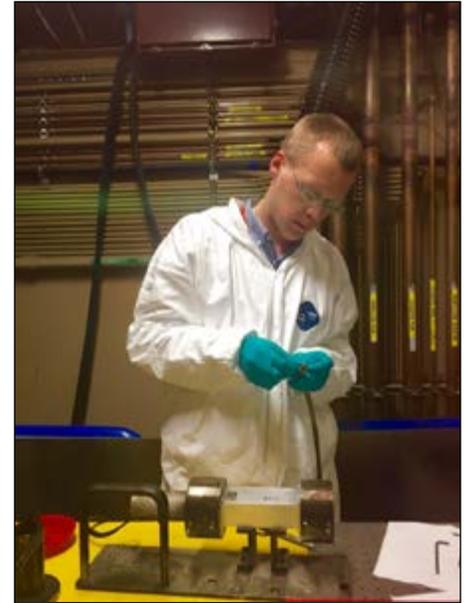
UTA graduate student Marc Schaeuble, at work in the Z pulsed power facility high-bay at Sandia National Laboratories, preparing a gas cell for a shot.

and Washington State University. ACE is featured in this issue of the *Stockpile Stewardship Quarterly* on page 9.

The University of Texas at Austin (UTA) Center for Astrophysical Plasma Properties (CAPP), led by Dr. Donald Winget, will focus on atomic and radiation physics of matter in a wide range of temperatures and densities. Although motivated by astrophysics, the Center will address problems of interest for stockpile stewardship, inertial confinement fusion, high energy density physics, and astrophysics. The CAPP team, comprised of the University of Texas at Austin and the University of Nevada, Reno, will use spectra at wavelengths from x-ray to optical to diagnose plasmas and to compare with observations of astrophysical objects and similar plasmas. This work will have a strong experimental emphasis but will incorporate a range of theorists and modelers for code validation. This will sharpen the scientific impact of the experiments and their contribution to NNSA.

Additional Center of Excellence awards will be made later in the fiscal year. ●