

ACTINIDE

RESEARCH QUARTERLY

LOS ALAMOS NATIONAL LABORATORY

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- ◆ RECONSTITUTION AS DETERRENCE
- ◆ THE HISTORY OF ACTINIDE SCIENCE
- ◆ ATOMICS



☀ ACTINIDE RESEARCH QUARTERLY

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Inside the next *ARQ*:

Highlights from Plutonium Futures—The Science Conference 2010, held in Keystone, Colorado, will be published in the upcoming Number 2 • 2011 issue of *Actinide Research Quarterly*.





Senior statesmen have joined with political leaders in calling for a recommitment to a world without nuclear weapons. Author Joe Martz took this photo of the senior diplomats (from left to right) former secretary of defense William Perry, former senator Sam Nunn, former secretary of state George Shultz, and former secretary of state Henry Kissinger.

RECONSTITUTION AS DETERRENCE ADVANTAGES AND CHALLENGES OF THE STRATEGY

Many people, including senior statesmen and political leaders, have suggested over the years that establishing and maintaining the capacity for reconstituting nuclear weapons may be a safer form of deterrence than retaining a large stockpile of weapons. In addition, in a widely read 2007 *Wall Street Journal* editorial, former secretary of defense William Perry joined former secretaries of state George Shultz and Henry Kissinger and former senator Sam Nunn in calling for a recommitment to achieving a world without nuclear weapons.

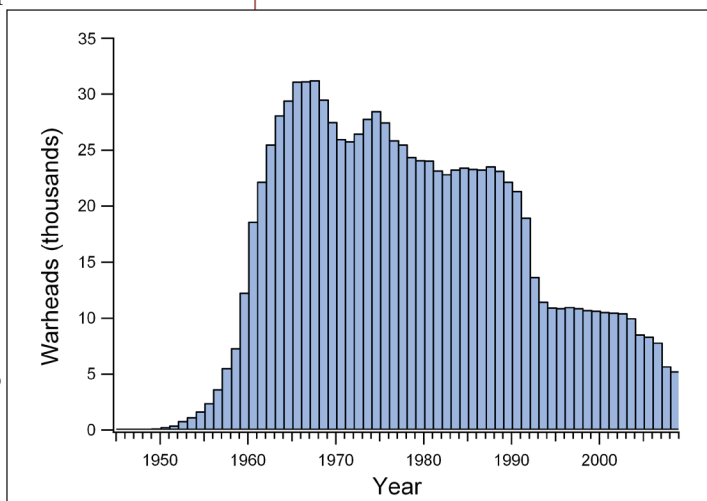
Two years later Secretary Shultz addressed the issue in the forward to *A World Without Nuclear Weapons: End-State Issues* by Sidney Drell and James Goodby. Shultz wrote: "The fact is nuclear deterrence is increasingly hazardous and decreasingly effective. We have to change our way of thinking about it ... including ways of stretching out time for decision making during a nuclear crisis and relying increasingly on an ability to reconstitute nuclear forces as a safer form of nuclear deterrence."

And as early as the 1980s, author Jonathan Schell was discussing what has become known as "capability-based deterrence" in his book *The Abolition*. "The capacity for retaliation would consist less and less of the possession of weapons and more and more of the capacity for rebuilding them, until, at the level of zero, that capacity would be all."

The current objective of capability-based deterrence is to accomplish two simultaneous goals: continue to preserve a strategic deterrent to aggression while enabling reductions

About the author: Joseph C. Martz of the Seaborg Institute contributed this article, which is based on a talk he gave last fall to the Los Alamos Committee on Arms Control and International Security. The topic is capability-based deterrence, which was the focus of Martz's recent research at Stanford University (see sidebar on page 9).

The number of U.S. nuclear weapons has decreased dramatically since 1965. By 2008 the stockpile was about 10 percent of what it was at its peak.



in nuclear weapons stockpiles. To do this, the nuclear weapons complex must demonstrate agility, capacity, confidence, security, and transparency.

Characteristics of an effective deterrent

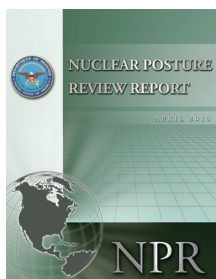
The United States' capability for producing nuclear weapons as a form of deterrent was robustly exercised during the Cold War to counter the threat of the Soviet Union. Looking forward, the desire is to preserve security in an environment of nuclear stockpile reductions, with a long-term vision of "Global Zero," in which the retention of deployed nuclear weapons isn't necessary to preserve the strategic security of the United States and its allies.

In support of the Global Zero vision, the Defense Department's 2010 *Nuclear Posture Review* (NPR) has embraced the idea that the reconstitution of nuclear forces can serve as a growing portion of deterrence in an environment of stockpile reductions. The Obama administration has backed up this policy decision with a recommendation to reinvest and revitalize the U.S. nuclear weapons infrastructure.

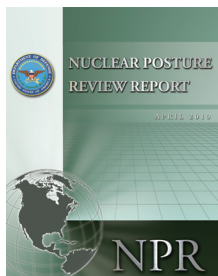
What remains to be decided are specific objectives and goals that address the strategy of further stockpile reductions and a move toward capability to preserve U.S. strategic security interests.

Some statesmen have begun to address this issue with specific proposals. Former Secretary Perry's "2020" vision establishes a concrete goal for stockpile reductions by the year 2020 (500 deployed weapons each for the United States and Russia), followed by a reexamination of the deterrence landscape. This proposal sets an intermediate goal that both preserves strategic security and makes substantial progress toward the Global Zero vision.

As the *Nuclear Posture Review* points out, the largest portion of the U.S. nuclear stockpile is not its deployed strategic forces; it is the reserve and backup forces, which are retained as a hedge against technical or geopolitical surprise. A vision of stockpile reductions can begin with a strategy that addresses possible reductions in these reserve forces. The *Nuclear Posture Review* has suggested that the ability to reconstitute nuclear forces can begin to augment, and eventually replace, the need to retain reserve and hedge forces.



"Implementation of the Stockpile Stewardship Program and the nuclear infrastructure investments recommended in the NPR will allow the United States to shift away from retaining large numbers of non-deployed warheads as a hedge against technical or geopolitical surprise, allowing major reductions in the nuclear stockpile. These investments are essential to facilitating reductions while sustaining deterrence under New START [Strategic Arms Reduction Treaty] and beyond." (NPR, page 30)



“Increased investments in the nuclear infrastructure and a highly skilled workforce are needed to ensure the long-term safety, security, and effectiveness of our nuclear arsenal and to support the full range of nuclear security work to include non-proliferation, nuclear forensics, counter-terrorism, emergency management, intelligence analysis and treaty verification. Such investments, over time, can reduce our reliance on large inventories of non-deployed warheads to deal with technical surprise, thereby allowing additional reductions in the U.S. nuclear stockpile and supporting our long-term path to zero. A revitalized infrastructure will also serve to reduce the number of warheads retained as a geopolitical hedge, by helping to dissuade potential competitors from believing they can permanently secure an advantage by deploying new nuclear capabilities.” (NPR, page 41)

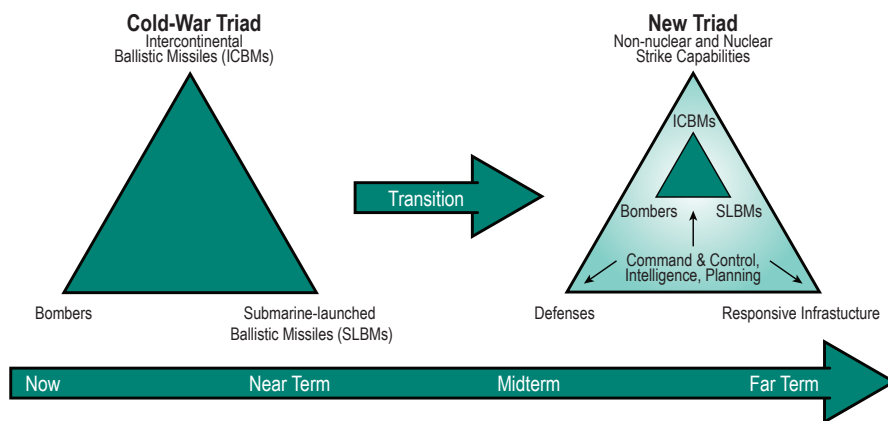
Building a capability-based deterrent

The essential questions for a capability-based deterrent are timing (agility) and capacity. There is no consensus on either of these issues at present, nor is there a ready answer to “how fast” and “how many” weapons or components should be reconstituted should the need arise.

Numerous studies in support of complex modernization have examined the structure of U.S. forces, the anticipated lifetime of various weapons and components, and the overall size of the deterrent. These studies have concluded that production capacities ranging from 50 to as high as 400 warheads per year are sufficient. These numbers are dramatically lower than the historic production capacities of the U.S. nuclear weapons complex, which produced as many as 8000 warheads per year in the late 1950s during the buildup of the Cold War.

Nonetheless, a production capacity of even 100 warheads per year is challenging, and key elements of the U.S. nuclear weapons complex are not presently configured to support this modest number.

The advent of long-range missiles gave rise to the nuclear triad for deterrence. This diagram, from the 2001 Nuclear Posture Review, shows the evolution of nuclear deterrence. Historically, each leg (delivery system) of the triad has unique abilities in support of deterrence. Intercontinental ballistic missiles (ICBMs) are land based and provide a visible counterforce target; submarine-launched ballistic missiles (SLBMs) provide survivable, second-strike assuredness; and bombers (with air-carried bombs and cruise missiles) are flexible, recallable, and ideal for “posturing” during a crisis. The historic triad remains as part of offense strike capabilities and is supplemented by both defense and infrastructure components.





Technicians inspect two Trident D5 missiles, which contain the W88 warhead. The first certified rebuilt pit for the W88 was produced at Los Alamos in 2006, almost eleven years after the program began. Much of this extended time was required to certify the newly built pit using the tools of stockpile stewardship.



A Trident missile is part of the SLBM “leg” of the nuclear deterrence triad. This photo shows a Trident D5 after launching from a British Royal Navy submarine.

These issues can be complex, and exact capacities will vary depending upon the urgency and need. For example, in a true national crisis, the “surge” capacity for pit (or other component) production could be substantially higher, given a willingness to modify safety and security rules. Furthermore, the required capacity for reconstitution in a geopolitical crisis could be substantially larger than the capacity calculated from stockpile size and lifetime considerations.

The question of agility (timing) is equally challenging to address. Recent experience in the weapons complex has demonstrated the capability to produce key components (such as pits) and complete weapons systems (life-extension programs) but arguably not on agile time frames.

For example, the first certified pit for the W88 was produced at Los Alamos in 2006, nearly eleven years after the program began. Much of this extended time was required to certify the newly built pit using the tools of stockpile stewardship (and most important, while conducting no further nuclear tests). If the United States is to rely upon reconstitution as a form of deterrence, the agility of the complex clearly must be improved.

Here, actinide science plays a crucial role in support of national security. The key elements of production and certification of pits intimately involve understanding the process–properties–performance relationship. This understanding encompasses physics performance and dynamic materials properties, as well as the engineering stability of the pit across the stockpile-to-target sequence, including critical performance in mechanical and chemical stability over decades. As recent experience in W88 pit production and in several life-extension programs has shown, there is a continuing need for further advancements in the scientific understanding and assessment of plutonium (and other actinides).

On an optimistic note, the complex has demonstrated substantial agility in several key programs. For example, the Reliable Replacement Warhead (RRW) feasibility study saw two independent teams conceive next-generation warhead designs and conduct substantial computational and experimental assessment of those designs in less than eighteen months.

Historically, the degree of design, computation, and assessment demonstrated by RRW would have taken four to five years. This shows the advantage of both modern engineering and modern design practices when applied to the nuclear weapons complex. It also shows the success and maturity of the Stockpile Stewardship Program in accomplishing its core function of assessing the safety, security, and effectiveness of the nuclear stockpile.

RRW exercised the front end of the design–certify–develop–manufacture cycle, which represents the spectrum required for a reconstitution strategy. Critically, the back end of this cycle, especially development and production, has been dormant for nearly two decades for many, if not most, of the materials and components in today’s weapons. Capability cannot exist by assertion alone; it must be exercised to be credible.

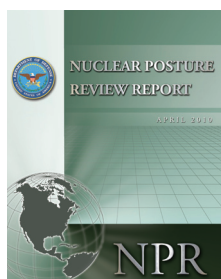
Thus, the environment today consists of four key elements. We have the recently ratified New START, which commits the United States and Russia to a

maximum of 1550 deployed warheads. The United States retains several thousand additional warheads as a reserve and hedge force. The administration and Congress have made a commitment to revitalize the nuclear weapons complex as a form of reconstitution-based deterrence. Finally, recent policies have embraced a path to further stockpile reductions, beginning with the just-released *Nuclear Posture Review*.

Strategy recommendations

An examination of these elements leads to a simple set of recommendations in the near term. We should formally and quantitatively adopt a strategy in which a growing demonstration of a capability-based deterrent begins to replace the reserve and hedge forces in the U.S. stockpile. Specifically, we should adopt a series of goals that when met allow the downsizing of U.S. reserve and hedge forces. These goals should be negotiated among all involved parties (NNSA, DOE, military, nuclear policy offices, Congress, and the nuclear weapons complex, with input from impacted allies) with specific dates, deliverables, resources, and associated numbers for reductions.

As the nuclear weapons complex demonstrates the ability to reconstitute specific—or functionally equivalent—weapons systems, the reserves for those weapons can be reduced. For example, a goal of delivering some number (a few dozen, perhaps) of a specific tail number (B61, W78, etc.) by some date would then trigger a reduction in the reserves for that weapon. The negotiation of these specifics will answer the questions of timing and capacity, resolving a key question in the formulation of a reconstitution strategy.



“Non-deployed warheads provide logistics spares, support the surveillance program, and hedge against technical or geopolitical surprise. ... Progress in restoring NNSA’s production infrastructure will allow these excess warheads to be retired along with other stockpile reductions planned over the next decade.” (NPR, page 38)

In support of this negotiation, the administration might appoint a group, cochaired by the U.S. Strategic Command and the NNSA, to develop specific goals, schedules, and resources. The timing of this recommendation is consistent with objectives in the *Nuclear Posture Review*, which states that replacing reserve and hedge forces with reconstitution capability is possible in the next decade.

Advantages of the strategy

The advantages of this approach are many. Foremost, establishing a robust capability to provide a strategic deterrent should the need arise is potentially more flexible than continuing the current U.S. strategy of maintaining



A W87 is part of the ICBM “leg” of the nuclear deterrence triad. This is a time-exposure shot of eight W87 Peacekeeper reentry vehicles launched from a single missile. The Air Force refers to these as reentry vehicles (RVs); the Navy refers to them as reentry bodies (RBs).



The B-61-11, often called an “air-carried” platform, is shown being loaded into a B-52 bomber.





Science-based stockpile stewardship has replaced nuclear testing to ensure a safe, reliable deterrent. Tools used include powerful simulations run on one of the world's fastest supercomputers, the IBM Roadrunner (above); studies of the effects of aging on weapons using an accelerated-aged plutonium alloy (right); and nonnuclear testing, such as the Reliable Replacement Warhead project's high-explosives hydrotest (far right).

Cold War-era weapons designed and built more than thirty years ago. Indeed, should a new threat emerge that requires a different balance of characteristics in the deterrent, the United States has few options available today to address this threat.

To cite one example, Russia has recently deployed a next-generation strategic warhead with terminal maneuverability on reentry. Such advances may require modifying or adapting U.S. forces to develop an adequate response. A robust capability provides this flexibility. More generally, the experience gained by the United States in its policy and leadership communities by addressing specific questions of timing, capacity, security, and confidence in constituting this capability will be critical as the country moves toward a future of fewer weapons and a more-capability-based deterrent.



Another advantage is the experience gained in trading reserve forces for reconstitution capability. In the longer term, it may be desirable to extend the contribution of a capability-based deterrent to provide functions that are currently served by deployed forces. This is a challenging assignment, and constraints on agility, capacity, survivability, confidence, and transparency will have a greater negative impact as more of our strategic security is vested in a capability.

The experience gained from an initial move from reserve forces to capability will be an essential steppingstone toward further transition in the U.S. nuclear force posture.

Yet another key advantage is the concrete revitalization of the nuclear weapons complex, especially the training and mentoring of the next generation of personnel and the concurrent advances in related scientific disciplines, notably actinide science. RRW is relevant here because it provided an opportunity for generational transfer of expertise.

The experiences in that study showed the irreplaceable nature of doing actual design work as opposed to focusing stockpile stewardship activities on assessment and certification. It is imperative to extend this experience to the development and manufacturing elements of the complex—and well before the loss of critical expertise makes rebuilding capability considerably more difficult.



RRW provided an additional lesson as well. It showed that the nuclear weapons complex responds best when it is given a specific assignment with concrete resources, milestones, dates, and the support of a broad spectrum of the community. A challenging assignment can serve to revitalize and motivate the national labs and production complex, just as RRW did for the nuclear weapons design enterprise.

Finally, the advantages of this approach in the international community are considerable. Establishing concrete goals and milestones for reducing reserve and hedge forces offers a powerful signal to the international community of our commitment to nonproliferation treaty objectives. Much of this can be accomplished unilaterally, with a powerful message to other nuclear states and the international community at large.

The transparency of these operations will be key in showing our security partners that our capability is robust and that their security is protected. Transparency will also play a role in showing that U.S. actions match our words with respect to the Global Zero vision as well as to other policy elements presented in the *Nuclear Posture Review* and other venues.

Experience gained with the international community in transitioning from stockpile numbers to a more-capability-based approach may form the basis for further arms-control efforts in which reserve forces and possibly even production capacity come under the umbrella of future agreements.

Trust gained among partners and the wider community may lay further groundwork for a greater transition to a capability-based approach in the distant future, helping to replace even-larger numbers of deployed forces.

Challenges of the strategy

The approach suggested here poses several challenges as well. The three most substantial are sustaining a commitment to a robust capability, maintaining confidence in this capability without additional nuclear testing, and addressing negative perceptions the international community may have of this capability if not convinced of the influence it can have on the nonproliferation and arms-control regimes.

A sustained commitment to the nuclear weapons complex is essential to ensure a robust capability over the long term. Over the last two decades, much of the capability in the nuclear weapons complex has eroded, and most of the production capacity has been closed or radically downsized. The need to revitalize the nuclear weapons complex has arisen due to this loss of capability and capacity.

Commitment is intrinsically a political and policy issue. Given the role that a revitalized complex will play in further stockpile reductions, and given a clear elucidation of a strategy and roadmap to Global Zero, the result of a cost-benefit analysis for investments in the nuclear weapons complex is compelling. Establishing the linkage between investments in capability and stockpile reductions may go a long way to ensuring this commitment.



One of the key questions for a capability-based deterrent is how the United States will be perceived by the rest of the world—allies as well as adversaries.





Confidence in a rebuilt stockpile in the absence of additional testing is another challenge. The Stockpile Stewardship Program has developed confidence in rebuilt and life-extended components, although the time frame has been less than agile in many examples. Execution of a large-scale project such as development of an agile reconstitution capability (the design–certify–develop–manufacture cycle) can be viewed as having three critical, interrelated components: scope, schedule, and resources.

Because scope is defined by process (the capacity and types of systems for reconstitution) and schedule is determined by agility constraints (several years, certainly not more than a decade), resources are a critical variable. Here, prior investments in the Stockpile Stewardship Program are crucial, as is the modernization of antiquated facilities, especially those that process nuclear material.

Confidence will come down to the breadth and scope of analysis for life-extended systems and, in some cases, improvements in the “performance margin” for certain system components during this process. Committing sufficient resources and support for the science-based tools of stewardship is absolutely essential to ensuring confidence in the absence of additional nuclear testing.

A final challenge is raised by the perceptions of the international community, perceptions that can be changed by the influence of this strategy on the nonproliferation and arms-control regimes. Viewed in isolation, establishing the capacity for reconstituting nuclear weapons may be seen as provocative. Conversely, when linked with substantial stockpile reductions and shown as part of a strategy for further advancing Global Zero objectives, reconstitution may be acceptable.

Joe Martz (below left) interviewed former secretary of defense William Perry in Palo Alto, California, last November. The interview will be included in a new video featured at Los Alamos’ Bradbury Science Museum.



Linkage and transparency are the key tools in this arena. During informal discussions the author had at Stanford University with a wide spectrum of international nuclear policy experts (including those from Sweden, Mongolia, the United Kingdom, India, Pakistan, and China), participants showed an interest in and acceptance of this approach if it is concretely tied to stockpile reductions. Formally establishing milestones that link reductions to development of reconstitution capability will powerfully serve this function.





MARTZ RETURNS AFTER STINT AS PERRY FELLOW IN INTERNATIONAL SECURITY

Los Alamos' Joseph C. Martz recently completed a one-year appointment as the inaugural William J. Perry Fellow in International Security at Stanford University. Martz, whose career at Los Alamos goes back twenty-seven years, is a nuclear materials scientist currently with the Seaborg Institute at Los Alamos. Among his areas of expertise are plutonium surface chemistry and metallurgy, including oxidation, dispersal mechanisms, and aging.

The Perry Fellowship was established in honor of former secretary of defense William J. Perry, an alumnus of Stanford University. Perry, along with former secretaries of state George Shultz and Henry Kissinger and former senator Sam Nunn, wrote an editorial in the *Wall Street Journal* in 2007 calling for a recommitment to achieving a world without nuclear weapons. The editorial received worldwide exposure, and President Barack Obama endorsed its objective in an April 2009 speech in Prague.

The fellowship expands upon the expertise available at Stanford's Center for International Security and Cooperation (CISAC), which has been at the forefront of nuclear policy and deterrence issues since its inception almost thirty years ago. CISAC's research on foreign nuclear weapons programs is among the most comprehensive and widely recognized in the world, and its current faculty includes former national laboratory directors Sig Hecker from Los Alamos, who serves as co-director, and Mike May from Livermore. Perry is currently the co-director of CISAC's Preventive Defense Project.

As the first Perry Fellow, Martz focused his research on technical questions surrounding the idea that the capabilities of the national laboratories and the nuclear weapons complex can serve as a growing part of deterrence.

"The opportunity to spend a year at Stanford working with Bill Perry, George Shultz, and Sig Hecker has been a career highlight," says Martz. "There is a hunger in the wider policy community for technical expertise and experience in nuclear weapons and nonproliferation. With the many developments in nuclear policy this past year, including the New START and the new *Nuclear Posture Review*, I really felt like a witness to history. My colleagues at Los Alamos should be enormously proud of their work. I saw firsthand how important the work of Los Alamos is in the policy community, and the appreciation and reverence for Los Alamos was palpable among the most senior statesmen."



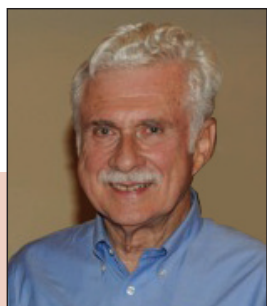


DECADES LATER: A FIELD THAT REMAINS “PARTLY UNDER CONSTRUCTION”

TRACING THE EVOLUTION OF ACTINIDE SCIENCE RESEARCH IN THE UNITED STATES

Many scientists entering a research field assume that the science has been funded steadily and has had broad public support for many decades. But the genesis of public support of science, the tradition of government support of basic research, and the maintenance of research support must not be taken for granted. Actinide science has a rich and dynamic history in the United States, but it is also an example of a research field that has seen government and public support wax and wane throughout the course of seventy years.

The field of actinide science was born with the discovery of neptunium and plutonium—within the memory span of some living scientists. The U.S. federal government has supported research on the physical, chemical, and nuclear properties of the actinides, with a focus on the transactinides, since the early days of World War II. Actinide research has been carried out under the auspices of the Department of Energy (DOE) and its predecessor agencies: beginning with the Uranium Committee in 1939 and the numerous groups that evolved into the Manhattan Project, the Atomic Energy Commission (AEC), and the Energy Research and Development Agency (ERDA).



About the author: Lester R. Morss began his scientific career in inorganic chemistry and radiochemistry by carrying out research on the actinide elements uranium through californium under Professor Burris B. Cunningham. He received a Ph.D. from the University of California, Berkeley, in 1969. After postdoctoral study with

James W. Cobble at Purdue University, he reached the rank of associate professor of chemistry at Rutgers University, performing research in synthetic inorganic chemistry and thermochemistry of transition elements.

He joined the Chemistry Division of Argonne National Laboratory in 1980, where he resumed his primary research focus on solid-state and thermochemistry of the transuranium elements. After reaching the rank of senior chemist at Argonne, he was elected a fellow of the American Association for the Advancement of Science and spent six months as an Alexander von Humboldt senior research scientist at the University of Hannover, Germany, in 1992.

Morss retired from Argonne in 2002 and then served until 2010 as program manager for Heavy Element Chemistry in the Office of Basic Energy Sciences of the Department of Energy. He is now an adjunct professor of chemistry at University of Maryland, College Park. He is co-editor, along with Jean Fuger and Norman Edelstein, of the recently published third and fourth editions of *The Chemistry of the Actinide and Transactinide Elements*.

The author acknowledges helpful comments from John Burnett, Norman Edelstein, Richard Haire, Robert Penneman, and Elliot Pierce.



The now-obscure growth and maturation of physical chemistry, of which actinide science is a part, is illuminated in a recent book, *Cathedrals of Science: The Personalities and Rivalries That Made Modern Chemistry*. The book's title is based on the preface to the 1923 textbook, *Thermodynamics and the Free Energy of Chemical Substances*, which reads in part:

"There are ancient cathedrals which, apart from their consecrated purpose, inspire solemnity and awe. Even the curious visitor speaks of serious things ... The labor of architects and artisans has been forgotten, the scaffolding erected for their toil has long since been removed, their mistakes have been erased, or have become hidden by the dust of centuries. ... But sometimes we enter such an edifice that is still partly under construction; then the sound of hammers ... enable[s] us to realize that great structures are but the result of giving to ordinary human effort a direction and a purpose."



Actinide science before World War II

European university laboratories advanced the field of nuclear science with early studies of the chemical properties of radioactive elements and the chemical effects of ionizing radiation. German chemist Martin Heinrich Klaproth discovered uranium in the 1780s, more than a century before French physicist Henri Becquerel discovered radioactivity in uranium minerals. Swedish chemist Jöns Jacob Berzelius discovered thorium in 1828. Actinium and protactinium are also found in nature because they have isotopes that are decay products of long-lived thorium and uranium ores; several of their isotopes were chemically separated by Marie Curie and other radiochemists working in France, Germany, and England between 1898 and 1909.

By 1941 chemical and physical properties of the first four actinide elements (actinium, thorium, protactinium, and uranium) were known, although neither the concept nor the terminology of an actinide series of elements had been expressed.

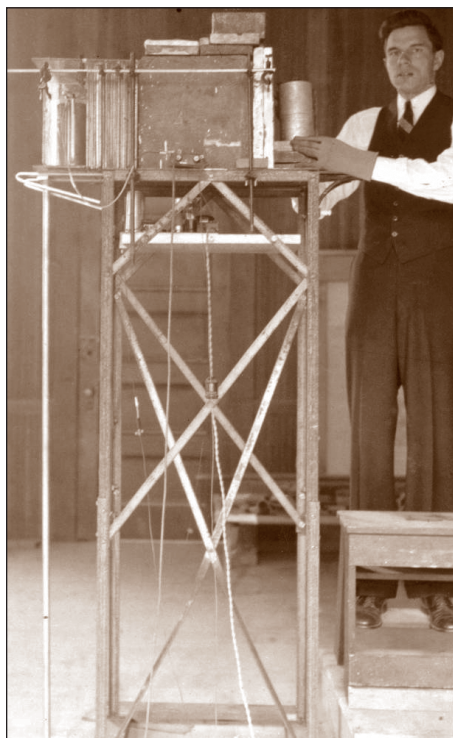
Nuclear science in the United States was practiced by a relatively small number of chemists. Theodore William Richards—an analytical chemist, not a radiochemist—made precise measurements that provided strong evidence that differences in the atomic weight of lead samples taken from different minerals (for example, pitchblende and thorite) were due to different isotopic ratios caused by radioactive decay. Richards was the first American to be awarded the Nobel Prize in chemistry, which he received in 1914.

Harold Urey—also not a radiochemist but a physical chemist—was inspired by a 1931 paper on differences in the atomic weights of hydrogen to search for a

Pioneers of nuclear science (clockwise from upper left): Marie and Pierre Curie; Henri Becquerel, who shared the 1903 Nobel Prize in physics with the Curies; Theodore William Richards, the first American to be awarded the Nobel Prize in chemistry; and G. N. Lewis, who mentored twenty Nobel Prize winners.

Lewis photo: Lawrence Berkeley National Laboratory





Glenn Seaborg was the quintessential actinide chemist. This photo from 1937 shows him with neutron-scattering apparatus in the East Hall at UC Berkeley.

Photo: Lawrence Berkeley National Laboratory

heavy hydrogen isotope, which he discovered and named deuterium. His studies of deuterium were an example of nuclear chemistry but not radiochemistry because neither deuterium nor heavy water involved radioactivity. He was awarded the Nobel Prize in chemistry in 1934.

Similarly, G. N. Lewis, already famous as a physical chemist but without a Nobel Prize, decided in 1935 to study the chemistry of heavy water and other deuterated compounds, perhaps as a “short and sure route to the Nobel Prize,” says Patrick Coffey in *Cathedrals of Science*. Although Lewis published twenty-six communications within less than two years on this topic, the Nobel Prize continued to elude him. He was nominated for the prize more than thirty times but never received it. He did, however, mentor twenty future Nobel Prize winners during his career.

Aristid von Grosse was a notable pre-World War II radiochemist who is considered by many to be the first U.S. actinide chemist. Educated in Germany, von Grosse came to the United States in 1930 and studied protactinium at Lindsay Light and Chemical Co. in West Chicago, Illinois, and at the University of Chicago in the 1930s. (Protactinium is a decay product of uranium.) Von Grosse developed techniques to recover and purify the element from uranium ores, isolated milligram amounts of the protactinium oxide Pa_2O_5 , reduced it to the metal, determined its atomic weight, and prepared several compounds.

Glenn Seaborg was the quintessential actinide chemist. He received his doctorate in 1937 from the University of California, Berkeley (UC Berkeley), under chemist George Gibson. Seaborg served as Lewis’s research assistant in generalizing the theory of acids and bases from the Arrhenius concept of protonic acids and from Lewis’s earlier concept of electron-pair acceptor “Lewis” acids. The result was a generalized acid-base concept in nonaqueous systems, organic chemistry, and catalysis. Seaborg then began to work in the field of nuclear chemistry. In 1939 Seaborg began a tenure-track career, first as an instructor and then as an assistant professor at Berkeley. He was thus able to begin an independent research career. The Los Alamos and Livermore Glenn T. Seaborg Institutes and the G.T. Seaborg Center at UC Berkeley/Lawrence Berkeley National Laboratory would later be named in his honor.

Seaborg recalled a Journal Club meeting (probably in January 1940) of the Physics Department at which an announcement was made about the Otto Hahn–Fritz Strassmann fission paper (“On the detection and characteristics of the alkaline earth metals formed by irradiation of uranium with neutrons,” published in *Naturwissenschaften* in 1939). Seaborg wrote, “Somebody got up and said, ‘You know, all of these transuranium elements ... are due to the splitting of uranium in half ...’ Before he had finished the sentence, I said to myself, ‘My God, how stupid we have been! Obviously, that should be the explanation.’” The fissionability of uranium-235 and the potential of a critical mass leading to a chain reaction and a nuclear explosion were tempered by the difficulty of separating the small concentration of uranium-235 from uranium-238 in natural uranium.

Meanwhile, Berkeley physicists Edwin McMillan and Philip Abelson had begun studies in nuclear chemistry. In 1937–39 they irradiated natural uranium with neutrons and succeeded in producing two radioisotopes, one with a half-life of 23 minutes and the other with a half-life of 2.3 days. McMillan identified the 23-minute isotope as uranium-239, previously identified by Austrian physicist Lise Meitner and others. Subsequently, in a few days' research during a May 1940 visit to Berkeley, Abelson discovered that uranium-239 decays by beta decay to a unique isotope: neptunium-239, the first isotope of a transuranium element.

It should be noted that Enrico Fermi and collaborators at the University of Rome carried out neutron bombardments of many elements, succeeding in inducing artificial radioactivity from many of them. Using careful radiochemical "carrier" chemistry, they ruled out most known elements as representing some of the artificial radioactivity and claimed discovery of transuranium elements. Fermi was awarded the 1938 Nobel Prize in physics in part for these discoveries, which turned out to be erroneous. What he thought were transuranium elements were subsequently found by Hahn and Meitner to be fission products. Fermi emigrated to the United States after receiving the Nobel Prize and extended his work in nuclear physics and reactor physics at Columbia University and the University of Chicago.

During the summer of 1940, McMillan tried unsuccessfully to identify the decay product of the 2.3-day isotope, neptunium-239. (The decay product is plutonium-239, but its long half-life, 24,110 years, prevented its detection at that time.) With permission of McMillan, who left Berkeley and moved to MIT to join the radar project, Seaborg and coworkers continued studying deuterium-irradiated uranium, discovering first (in late 1940) the much more radioactive plutonium-238 (half-life 87.7 years) and, early in 1941, the longer-lived plutonium-239 and its fissionability.

None of this pre-World War II chemistry research on actinide elements was supported by government grants or contracts. There were no such grants or contracts until 1940, except for applied research related to military needs. Other than the War and Navy Departments, only the National Bureau of Standards and the Department of Agriculture had established research programs. But in 1939 nuclear scientists took the first steps to establishing the tradition of government support for research applied to national needs.

Evolution of the Manhattan Project

In the years immediately preceding the United States' entry into World War II, while researchers investigated the new science of fission, decisions were being made in Washington, D.C., that would evolve into a full-scale bomb project—the Manhattan Project.

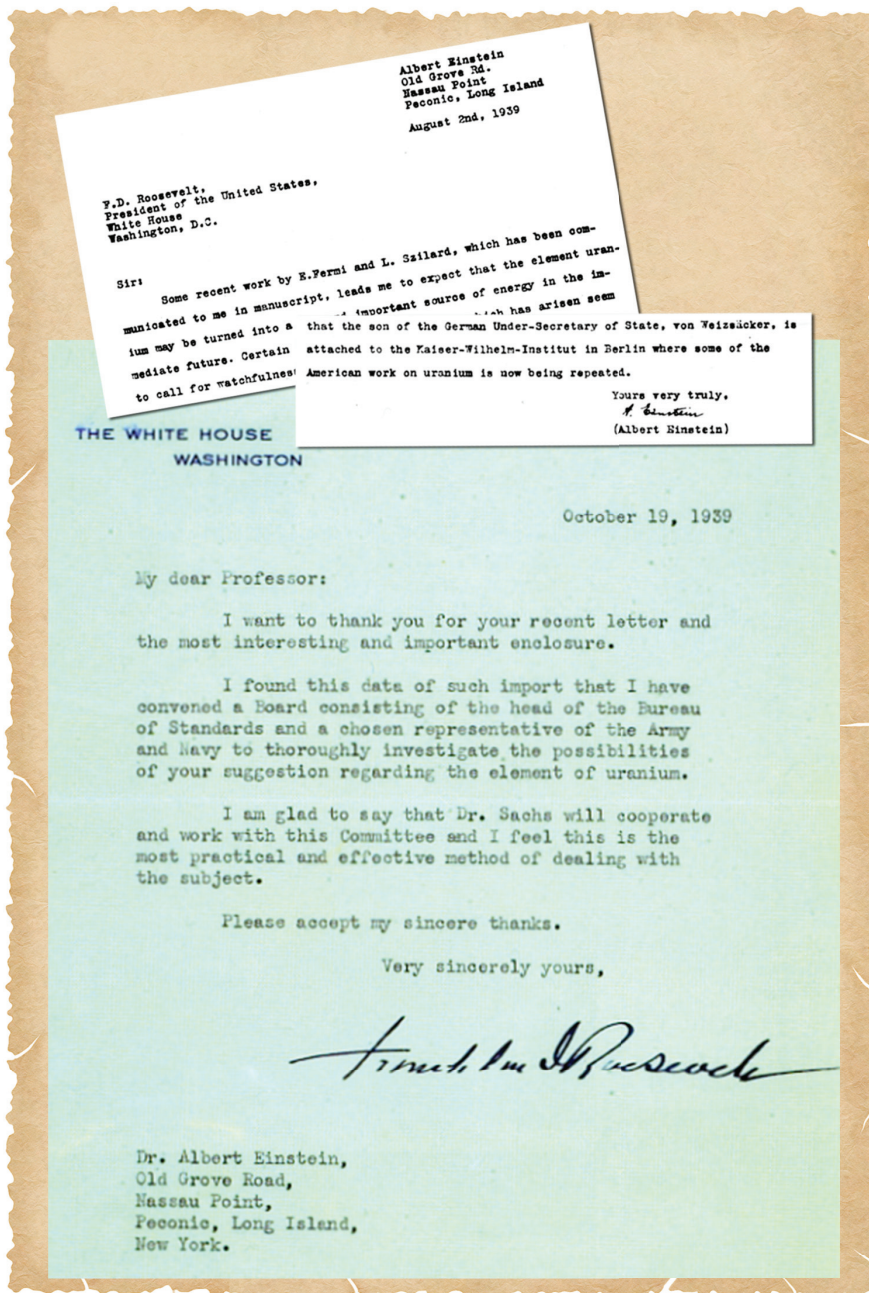
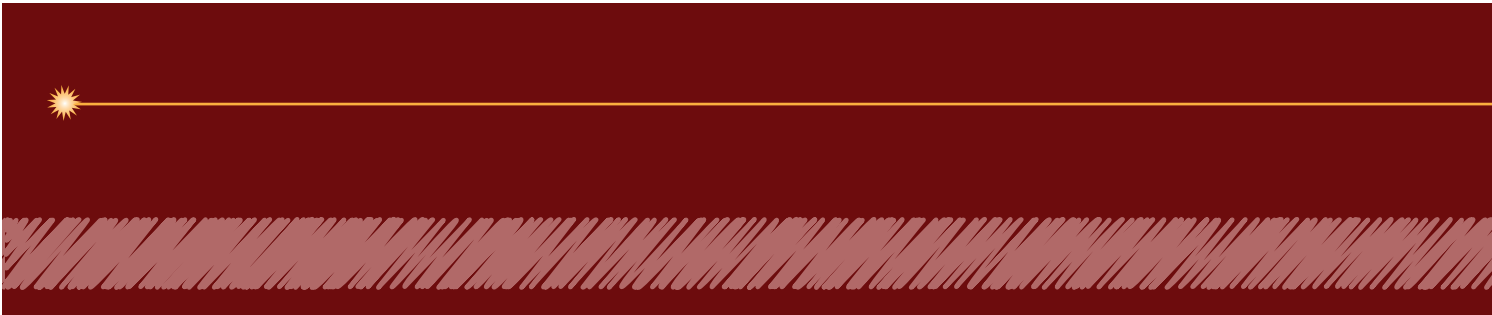
In 1939, in response to the famous letter of Albert Einstein warning President Franklin Roosevelt of the military potential of nuclear fission, Roosevelt established the Advisory Committee on Uranium to consider the



Edwin McMillan in the lab in 1940, the year he discovered neptunium. He and Glenn Seaborg would share the 1951 Nobel Prize in chemistry for "their discoveries in the chemistry of the transuranium elements."

Photo: Lawrence Berkeley National Laboratory





feasibility of an atomic bomb. He appointed Lyman J. Briggs, director of the National Bureau of Standards, to head the advisory committee, which included both military and civilian members. The committee met for the first time on October 21, 1939, and several months later recommended that the government fund research—\$6,000 was budgeted for research on fission chain reactions and isotope separation.

In June 1940 the Uranium Committee was transferred to the newly created National Defense Research Committee (NDRC), chaired by Vannevar Bush, an MIT physicist and president of the Carnegie Institution. Bush would become one of the most influential forces in the establishment of not only the nascent Manhattan Project but also early U.S. atomic-energy policy.

With Roosevelt's approval, Bush reorganized the Uranium Committee into a strictly scientific committee and eliminated the military membership. In June 1941 Roosevelt appointed Bush as director of another new agency: the Office of Scientific Research and Development (OSRD). Bush reorganized the Uranium Committee into the Section on Uranium, code name S-1, at which time jurisdiction for it was transferred from the NDRC to the OSRD. Bush now had responsibility for all fission research, and Briggs reported to Bush.

James B. Conant, a chemist and president of Harvard University, replaced Bush at the NDRC. While the NDRC technically still existed after the creation of the OSRD, its authority was reduced from actually funding research to serving as an advisory body to the OSRD. The NDRC would cease to exist after its last meeting, in January 1947.

In early July 1941 the British MAUD Committee issued a report concluding that a uranium bomb was feasible. (MAUD is

In 1939 Albert Einstein wrote President Roosevelt a letter (top) warning him of the military potential of nuclear fission. Roosevelt's reply to Einstein (above) says in part: "I found this data of such import that I have convened a Board . . . to thoroughly investigate the possibilities of your suggestion regarding the element of uranium." Thus was born the Advisory Committee on Uranium, with Lyman J. Briggs at its helm.





Vannevar Bush became one of the most influential forces in the establishment of Manhattan Project and early U.S. atomic-energy policy.



The S-1 Executive Committee in 1942. From left to right: Harold Urey, E. O. Lawrence, James Conant, Lyman Briggs, Eger Murphree, and Arthur Compton.

often assumed to be an acronym, but it was actually the name of the governess of Danish physicist Niels Bohr's children.) A report by the U.S. National Academy of Sciences later that year agreed with the MAUD Committee's conclusion. In December 1941 Bush organized a meeting to accelerate research into uranium-235. Arthur Holly Compton, Nobel Prize winner and physics professor at the University of Chicago, was in charge of the project to investigate gaseous diffusion and electromagnetic enrichment of uranium-235. Urey headed heavy water and isotope separation research, and Fermi headed theoretical studies. The S-1 project now focused on developing an atomic bomb.

In June 1942 Bush dissolved the original S-1 and created the S-1 Executive Committee, whose members included Conant (chairman), Briggs, Compton, Urey, E.O. Lawrence (winner of the Nobel Prize in physics in 1939 for his work on the cyclotron), and Eger Murphree (a chemist with Standard Oil). Cooperation between the OSRD and the Army was strengthened, and the project was put under the management of the U.S. Army Corps of Engineers.

On August 13, 1942, the Manhattan Project (formally called the Manhattan Engineer District because its first offices were in New York City) was created, and on September 17, 1942, General Leslie R. Groves assumed command. Three primary secret research and production sites were established: Site W at the Hanford Site in eastern Washington state for plutonium production; Site X at Oak Ridge, Tennessee, for uranium isotope separation; and Site Y in Los Alamos, New Mexico, for bomb design. Berkeley theoretical physicist J. Robert Oppenheimer was named scientific director at Los Alamos. By May 1943 the Army had assumed full control over OSRD's research projects, and the S-1 Executive Committee became inactive. The Manhattan Project would later involve more than 30 sites, including universities, and 130,000 people.



Site W: Hanford B-reactor area (top). Site X: Oak Ridge Y-12 shift change (center). Site Y: Los Alamos main technical area (above).





Glenn Seaborg looking at the first pure plutonium produced at the University of Chicago Met Lab, 1942.

Photo: Lawrence Berkeley National Laboratory

Actinide science during World War II

While Washington was consolidating the institutional structure that would lead to development of the atomic bomb, scientists were making major discoveries.

NDRC leaders realized that the key to a uranium fission weapon was separating the rare isotope uranium-235 from the more abundant uranium-238. Processes to accomplish isotope separation were then, and still are, physical processes (such as diffusion or centrifugation) rather than chemical processes. When Seaborg succeeded in proving that plutonium-239 was fissionable, an alternative “chemical” route to a fission weapon became possible.

The first fission reactor Fermi constructed at the University of Chicago might have had sufficient neutron flux to produce multigram quantities of plutonium-239. Plutonium could be separated from uranium-238 by a chemical rather than a physical process. In 1941 Compton chaired the National Academy of Sciences Committee to Evaluate Use of Atomic Energy in War. Surprisingly, it was Compton—a physicist—who favored pursuing the plutonium-239 “chemical” route to nuclear fission, whereas Conant—a chemist—preferred only the uranium-235 “physical” route. Compton’s influence led directly to the plutonium chemistry research program at the University of Chicago Metallurgical Laboratory (Met Lab), to the Oak Ridge and Hanford reactors, and to the Trinity Site and Nagasaki plutonium nuclear explosions.

Actinide chemistry research that focused on a plutonium bomb option was initiated in the spring of 1942, primarily in Section C-1 at the Met Lab under Seaborg’s leadership. A secondary site was at the UC Berkeley Chemistry Department under Wendell Latimer, Robert Connick, Leo Brewer, and John Gofman. Almost all of this research focused on plutonium chemistry. Plutonium metallurgy was carried out first at the Met Lab and expanded at Los Alamos.

Chemist Frank H. Spedding led important wartime research at Iowa State College (now Ames Laboratory of Iowa State University). Spedding developed technology to make high-purity uranium metal in sufficient quantities for reactors. He also developed ion-exchange separation of rare-earth elements, a technique that would later be applied to separate and chemically characterize all transplutonium actinide elements.

Although elements heavier than plutonium were in a strict sense peripheral to the mission of the Manhattan Project, both nuclear physicists and chemists were aware that reactor production of plutonium would also result in elements of higher atomic number, created by the beta decay of plutonium isotopes (for example, plutonium-241 forming americium-241 and plutonium-243 forming americium-243), followed by additional neutron capture. These transplutonium isotopes would degrade weapon performance, so their physics and chemistry had to be studied. The first transplutonium isotopes were actually prepared in the summer of 1944 by alpha-particle bombardment of plutonium-239 to make curium-242 and, later that year, by neutron bombardment of plutonium-239 to make americium-241. However, the isotopes’ isolation and

identification remained elusive because no one anticipated that their +3 ions would be the most stable.

During the Manhattan Project years, Seaborg hypothesized that these elements might form an “actinide series” with stable +3 ions. Seaborg enunciated the “actinide concept” in classified Met Lab papers in 1944. In talks and papers, he frequently mentioned that, for example, “when I showed [the actinide concept] to some world-renowned inorganic chemists, I was advised not to publish it—such an act would ‘ruin my scientific reputation.’”

Seaborg’s first public description of the transplutonium elements was on a “Quiz Kids” radio program in Chicago on November 11, 1945, a few days before his paper on the subject was presented at an American Chemical Society symposium and immediately published. The phrase “heavy-element chemistry” traces its origin to studies of “heavy elements” or “heavy isotopes” during and immediately after World War II. (For more on Seaborg, see *ARQ*, 2nd Quarter, 2009.)

The frantic pace of wartime actinide research culminated in the production of three atomic bombs: one uranium gun-type assembly, in which a subcritical mass of uranium-235 is shot at another subcritical mass of fissile material, and two plutonium-triggered implosion devices, in which a core of plutonium is compressed to critical mass by a high-explosive charge.

The scientists had little doubt that the uranium gun assembly would work but were concerned about the implosion concept. For that reason, they chose to test the plutonium device, nicknamed “The Gadget”—the world’s first atomic bomb—at Trinity Site in Alamogordo, New Mexico, on July 16, 1945. (A test of the implosion device rather than the gun-type one was also prudent because there was much more plutonium available than uranium-235.) The implosion test was a success. The first nuclear device used as a weapon, “Little Boy,” was the uranium-based bomb, dropped on Hiroshima, Japan, on August 6, 1945. Three days after that, the second plutonium weapon, “Fat Man,” was exploded over Nagasaki, Japan.

Transuranium science began in the Soviet Union in 1944 with tracer amounts of neptunium and plutonium isolated from neutron and cyclotron bombardment of uranium-238. Transuranium science continued with reactor-produced samples in the Soviet Union and Britain after 1945, and later in other countries as well.

J. Robert Oppenheimer (center, in porkpie hat), Gen. Leslie Groves (to Oppenheimer’s left), and others inspect the remains of the tower at Trinity ground zero, September 1945.



Above left: Sgt. Herbert Lehr delivers part of the Gadget’s plutonium core in its shock-mounted carrying case to the assembly room at the McDonald Ranch house, Trinity Site. Above right: The Gadget at the base of the test tower, Trinity Site.



The Trinity Test, 9.0 seconds after detonation.





President Truman, with several senators looking on, signs the Atomic Energy Act of 1946, which established the U.S. Atomic Energy Commission.

Postwar actinide science

The Atomic Energy Act was introduced in Congress on December 20, 1945, during a time when there was much debate (mostly out of public view) over whether atomic energy should be under military or civilian control. The bill established civilian control, with many restrictions on dissemination of information, even to U.S. wartime allies. It was signed by President Harry Truman on August 1, 1946, and became law on January 1, 1947. Manhattan Project assets were transferred to the new Atomic Energy Commission (AEC) at midnight on December 31, 1946. The AEC exercised governmental control over military, regulatory, and developmental aspects of atomic energy until 1975 when the agency was disestablished.

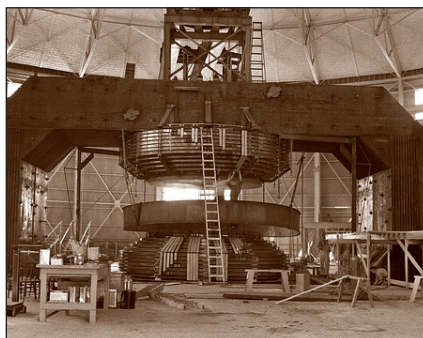
The AEC was formally led by five commissioners and supported by technical management led by a general manager. The first AEC general manager was Carroll L. Wilson, a 1932 MIT graduate, who was nominated by Truman (and subsequently confirmed by the Senate) even though he had little more than a decade of MIT and government management experience. The AEC's first director of research was James B. Fisk, a colleague of Wilson's. Fisk had taught physics at MIT and came to the AEC in 1947, after having served as wartime director of research at Bell Laboratories. Although at the AEC only until August 1948, Fisk initiated research in high-energy accelerators and expanded support of science at universities.

Meanwhile, the first AEC director of biology and medicine, Shields Warren, was able to initiate basic research in biological sciences because the Division of Biology and Medicine was parallel to, rather than under, the Division of Research. Warren had been chief pathologist at the New England Deaconess Hospital and professor of pathology at Harvard Medical School.

The second AEC director of research, Kenneth Pitzer, served from January 1949 through June 1951. Pitzer came from the chemistry faculty at UC Berkeley and returned there after his AEC tenure. He initiated AEC support for physical science research at the national laboratories and guided a transition from university contracts with Office of Naval Research interim support to those with AEC support.

The AEC's authority to issue research contracts outside the national laboratory system was tenuous: its legal staff concluded that Pitzer could legally participate in evaluating and selecting research projects as long as the Commission determined the total allocation for such research. However, in December 1950, at the height of the Korean War, Pitzer "believed that the Commission could take a more daring approach" to focus AEC research on applied research on military topics (as noted in *A History of the United States Atomic Energy Commission*, by Richard Hewlett and Oscar Anderson), clear scientists for classified research, and take steps so that "universities should be prepared to undertake classified research."

By law, Congress limited research to reactor physics, metallurgy, and related reactor science and to weapons development. Almost all research was carried out



at the national laboratories: first Argonne National Laboratory (the first national laboratory, chartered in 1946 and arising out of the Met Lab) and then Oak Ridge. Thus, postwar actinide science suffered from the exodus of many Manhattan Project scientists back to universities, where they could not seek AEC support. Robert Penneman, leader of the actinide group at Los Alamos Scientific Laboratory, was able to initiate americium chemistry there with the laboratory director's informal approval but without the AEC's formal approval. ("Scientific" was added to Los Alamos Laboratory's name in 1947, and in 1980 the name changed again to Los Alamos National Laboratory.)

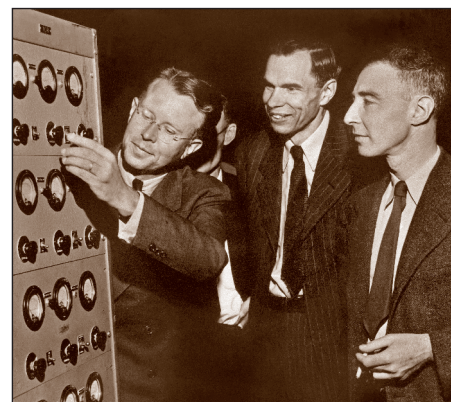
The Radiation Laboratory at Berkeley was established before World War II with private and university support; it became a federal laboratory in 1942 and an independent national laboratory in 1959. Actinide chemistry research there was led by Seaborg and Burriss Cunningham, who both returned to Berkeley in 1946 after leaving the Met Lab.

Seaborg served on the first General Advisory Committee of the AEC from January 1947 to August 1950, along with chairman Oppenheimer; Fermi; Conant; Isidor Rabi, a physicist and Nobel laureate; and Lee A. DuBridge, a physicist and founding director of the Radiation Laboratory at MIT. The General Advisory Committee advised the AEC to initiate a program of support for basic research in U.S. universities and colleges. The Atomic Energy Act did not permit issuing contracts or grants for such research; nevertheless, Pitzer was able to initiate a few non-national-laboratory contracts.

The National Science Foundation, proposed by Bush in 1945 in the influential report "Science: The Endless Frontier" and finally enacted into law in 1950, provided a model for independent support of scientific research based on peer-reviewed proposals from researchers rather than directed-research contracts awarded to national laboratories.

The '50s: Focus on the national laboratories

President Dwight Eisenhower's Atoms for Peace address to the United Nations General Assembly in December 1953 and the resulting Atoms for Peace program removed the cloak of secrecy from much basic actinide research. The Atomic Energy Act was modified in 1954 to permit a limited number of



Clockwise from upper left: Construction of the 184-inch cyclotron at U.C. Berkeley's Rad Lab began in 1941. The magnet yoke was set in place and the building erected around it. E. O. Lawrence and his staff pose with the magnet at the cyclotron, which was converted from a calutron to a synchrocyclotron after the war. Lawrence (left), Glenn Seaborg (center), and J. Robert Oppenheimer at the controls to the magnet in early 1946, while it was being converted from its wartime use.

Photos: Lawrence Berkeley National Laboratory





President Eisenhower delivers his Atoms for Peace address to the United Nations General Assembly, December 8, 1953.

From left to right: William Carnall spent his scientific career at Argonne National Laboratory, where he pioneered work in interpreting lanthanide spectra in solutions. After his death in 2003, the Handbook on the Physics and Chemistry of Rare Earths, Volume 37, was dedicated to him. Albert Ghiorso was a co-discoverer of a dozen elements, more than were discovered by anyone else. His research career spanned more than five decades, most of which was spent at Lawrence Berkeley Laboratory. Robert Penneman was leader of the actinide group at Los Alamos Scientific Laboratory. The group was one of only a few that carried out most of the pioneering transplutonium research in the United States. Gregory Choppin, a co-discoverer of mendelevium while at Lawrence Berkeley Laboratory, continued his actinide research at Florida State University into the twenty-first century.

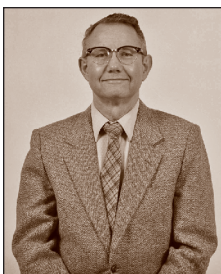
level-of-effort contract awards to outside organizations (for example, academic institutions). The modified act did not require merit (peer) review of proposals, although the AEC followed the National Science Foundation example by requesting proposals from universities and subjecting them to written merit review.

Among the scientists involved in pioneering research projects at Argonne National Laboratory, Paul Fields carried out nuclear and atomic spectroscopy, Joseph Katz studied actinide oxides and fluorides, Dieter Gruen studied molten salts, and Sherman Fried and Leonard Katzin synthesized new actinide compounds. Fields led the Argonne heavy-element group for many years, then served as Chemistry Division director in the 1960s and 1970s.

At the University of California Lawrence Radiation Laboratory (LRL), Seaborg continued nuclear chemistry research that led to the discoveries of berkelium (1949), californium (1950), einsteinium (1952), fermium (1953), mendelevium (1955), and nobelium (1958) and determination of their nuclear and chemical properties. These elements were separated as tripositive ions by cation exchange by Seaborg and colleagues, in particular Gregory R. Choppin, who continued his actinide research at Florida State University into the twenty-first century.

Seaborg's colleague Albert Ghiorso was first author on the papers that announced the discoveries of einsteinium, fermium, mendelevium, and nobelium, as well as that of lawrencium in 1961, when Seaborg was chairman of the AEC. Seaborg chaired the session "Heavy Element Chemistry" at the first Conference on the Peaceful Uses of Atomic Energy in Geneva in 1955. He gave a plenary lecture, "Recent Developments in the Field of Transplutonium Chemistry," at the second conference in 1958. The vast scope of these two conferences was manifested by the publication of research papers in twenty-six and thirty-two volumes, respectively.

Seaborg was influential in the initiation of the U.S. National Transplutonium Production Program that led to construction of the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. As americium and heavier actinide



William Carnall



Albert Ghiorso



Robert Penneman



Gregory Choppin



metals and compounds became available in microgram or larger quantities, Cunningham pursued microchemical syntheses of these actinides to determine thermodynamic, magnetic, spectroscopic, and electrochemical properties.

During this and adjacent decades, the Cunningham group at Berkeley, the Penneman group at Los Alamos, and the Fields–Carnall

group at Argonne carried out most of the pioneering transplutonium research in the United States. The Los Alamos actinide program, led by Penneman, characterized americium chemistry both in solids and solution. (For more on Penneman and the history of isotope chemistry at Los Alamos, see *ARQ*, 2nd/3rd Quarters, 2010.)

Eisenhower appointed Seaborg to the President’s Science Advisory Council in January 1959. The Advisory Council commissioned a study of the interactions among U.S. funding agencies and institutions that carried out basic research. Seaborg chaired the committee that conducted the study. The final report, “Scientific Progress, the Universities, and the Federal Government,” known as the “Seaborg Report,” made an immediate impression on Eisenhower in the final month of his presidency and had long-lasting influence.

Seaborg wrote, “Perhaps the report’s most famous recommendation was ... that basic research and the education of scientists go best together as inseparable functions of universities [and] that federal support for basic research and graduate education should be continued and flexibly increased, so as to support excellence where it already exists and to encourage new centers of outstanding work.”

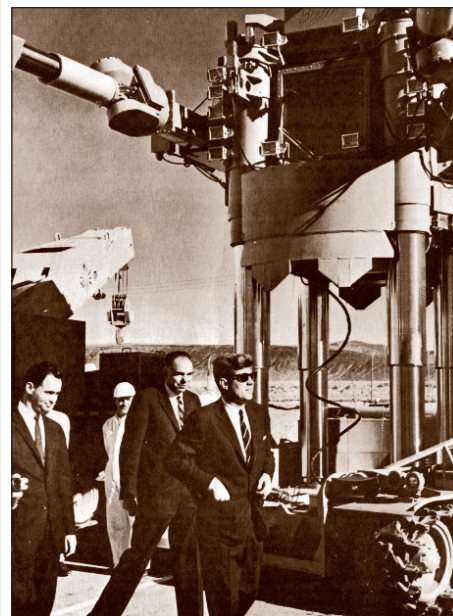
The '60s: The golden post-Sputnik years

Seaborg, who served as chancellor of UC Berkeley from 1958 through 1961, was nominated by President-elect John F. Kennedy to succeed John McCone as chairman of the AEC. He served in that position for ten years (1961–1971), being reappointed by Presidents Lyndon Johnson and Richard Nixon. During McCone’s tenure, the AEC expanded its role in producing nuclear materials, in particular, long-lived isotopes of the transuranium elements curium through fermium for research studies. Seaborg was instrumental in negotiating both international agreements on peaceful uses of atomic energy and the limited nuclear test ban treaty with the Soviet Union. He served as head of the U.S. delegation to the fourth United Nations Conference on the Peaceful Uses of Atomic Energy in Geneva in 1961.

Alexander VanDyken was assistant director for Chemistry Programs, Division of Research, at the AEC in the 1960s. In late 1964 the Transplutonium Program Committee was officially formed as an advisory body to the director of

The High Flux Isotope Reactor at Oak Ridge, completed in 1968, was commissioned to isolate and purify transplutonium isotopes from the reactor’s irradiation targets. At peak production levels in the 1980s, two fuel-rod-separation campaigns were conducted annually, producing berkelium-249, californium-252, einsteinium-254, and fermium-257.

President Kennedy and Glenn Seaborg tour the Nuclear Rocket Development Site at the Nevada Test Site in December 1962. As president-elect, Kennedy nominated Seaborg as chairman of the Atomic Energy Commission, a position he held for ten years.





In an article in the January 8, 1968, issue of Chemical & Engineering News, nuclear chemistry pioneers recalled the first weighing of plutonium. Featured on the cover were (from left to right) Louis Werner, Glenn Seaborg, Burris Cunningham, and Michael Cefola. This is a scan of the author's personal copy of the magazine.



the Division of Research to allocate Radiochemical Engineering Development Center products to U.S. national laboratories and to oversee the research carried out with them. VanDyken served as chairman.

Heavy-element chemistry continued at Berkeley, led by Cunningham, who in the late 1950s helped develop ultra-microchemistry using single cation exchange resin beads. This technique led to synthesis of pure compounds of berkelium-249, californium-249, and einsteinium-253 in microgram or smaller amounts by Cunningham and his students, followed by characterization of the physical and chemical properties of these compounds. The first research with products from the High Flux Isotope Reactor (HFIR)–Transuranium Element Processing Plant (TRU) would be carried out at Berkeley and Oak Ridge in 1967.

Seaborg had proposed Oak Ridge's HFIR in 1957. The AEC authorized it in 1958, and construction was completed in 1964. The companion TRU was authorized in 1958 and completed in 1965; it was later renamed the Radiochemical Engineering Development Center. The facility, which included heavily shielded gloveboxes and "caves," was commissioned to isolate and purify transplutonium isotopes from other components of the reactor's irradiation targets.

At the peak HFIR production level in the 1980s, two fuel-rod separation campaigns were conducted annually, producing about 50 milligrams of berkelium-249, 500 milligrams of californium-252, 3 micrograms of einsteinium-254, and 1 picogram of fermium-257. Heavier actinide and transactinide isotopes cannot be produced by neutron irradiation in reactors. Alfred Chetham-Strode, and later O. Lewin Keller, Jr., led Oak Ridge's actinide chemistry program. Inorganic chemists Russ Baybarz and Richard G. Haire advanced the understanding of actinides in low oxidation states and in colloids.

Haire led research thrusts in heavy actinide metal, oxide, and halide thermodynamic systematics, especially under high pressure. He also expanded upon Cunningham's ingenious syntheses of transplutonium materials at the milligram and microgram scale, coupling the syntheses of pure materials to measure and interpret their properties by systematic studies as a function of atomic number, f-electron configuration, temperature, and high pressure. Haire fostered collaborations with domestic and foreign laboratories, especially with the Institute for Transuranium Elements, Karlsruhe, Germany.

Argonne opened its "hot" laboratories (M-Wing of Building 200) in 1963, with hot cells for remote-handled high-level spent fuel examination and separations and fiberglass gloveboxes and hoods for f-element solid-state chemistry, spectroscopy, and solvent extraction separations.

At the Savannah River Site, Clark H. Ice, who later became director of Savannah River Laboratory, was influential in encouraging the separation of transplutonium isotopes, in particular curium-244 and californium-252, from Savannah River reactor targets before the startup of Oak Ridge's HFIR. David Karraker was the chemist most responsible for transuranium research there, carrying out magnetic susceptibility measurements of



americium compounds and synthesis of organoactinide compounds.

Among the few university researchers in actinide chemistry, Choppin, who was co-discoverer of element 101 (mendelevium) at Berkeley, carried out coordination chemistry and thermodynamic measurements at Florida State University from 1956 through 2008. James W. Cobble of Purdue University had a contract for thermodynamic studies that included uranium, neptunium, and plutonium. Joseph R. Peterson of the University of Tennessee had a contract for studies of transplutonium compounds from 1969 through 2000.



The '70s and '80s: Transition to DOE

AEC research activities that eventually became the Office of Basic Energy Sciences (BES) were originally in the Division of Research. In December 1971 the Division of Research was renamed the AEC Division of Physical Research to help distinguish it from an expanding program in biological, health, and medical research. The AEC became part of the new Energy Research and Development Administration (ERDA) as a result of the Energy Reorganization Act of 1974.

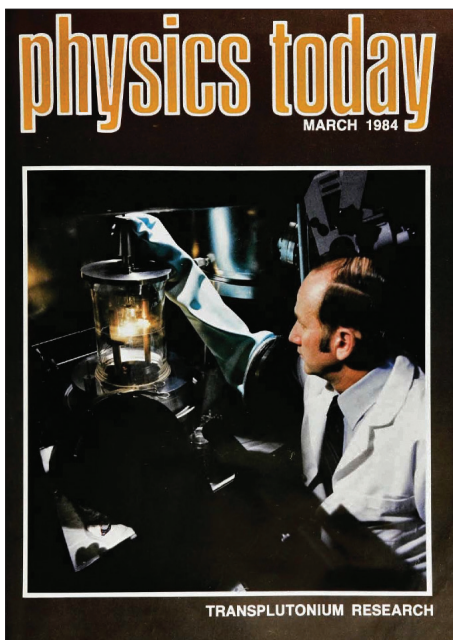
In 1975 Congress created the Nuclear Regulatory Commission (NRC) to oversee the nuclear power industry and other civilian uses of nuclear energy. The NRC was independent of ERDA, which administered energy research and development, including nuclear power. The AEC's weapons program was incorporated into ERDA. In 1977 ERDA and the energy programs from a number of other federal agencies (not including the National Science Foundation) were brought into the new cabinet-level Department of Energy (DOE). Major programs that were brought from other agencies into the new DOE included solar energy, energy efficiency, and fossil energy.

Although the broadened focus of this new Cabinet-level energy agency did not in principle undermine research strength from traditional AEC areas, heavy-element chemistry stagnated during the 1970s and 1980s as its Cold War justification began to wane, as nuclear energy suffered environmental stresses from the lack of a waste repository and from the nuclear accidents at Three Mile Island and Chernobyl, and as the cutting-edge excitement of new-element discovery required exotic and exhausting efforts to synthesize a few atoms of short-lived isotopes.

ERDA became part of DOE as a result of the Department of Energy Organization Act of 1977. As part of the formation of DOE, heavy-element chemistry and other chemical and material science research programs became part of the Office of Energy Research on October 1, 1977. The Heavy-Element

Glenn Seaborg (far left) as chairman of the Atomic Energy Commission, asked President Nixon (second from left) to present a special Atomic Pioneers Award to (center from left to right) Vannevar Bush, Gen. Leslie Groves, and James Conant for their service in running the Manhattan Project.





Oak Ridge's Richard Haire was featured on the cover of Physics Today in 1984 to illustrate an article on transplutonium research authored by O. Lewin Keller Jr., Darleane Hoffman, Robert Penneman, and Gregory Choppin. Haire is shown removing a spent fuel element from a pressure vessel at the High Flux Isotope Reactor.

Chemistry program was initially part of the Office of Nuclear Energy but was transferred without a change in focus to the Office of Basic Energy Sciences in 1983. Both offices were part of the DOE Office of Energy Research, which was renamed the Office of Science in 1998.

Elliot Pierce directed the Office of Chemical Sciences from 1973 through 1986. John Burnett was the program manager of the heavy-element chemistry program from 1969 through 1996. Pierce guided the program to separate and utilize the transplutonium isotopes that were generated in the HFIR reactor at Oak Ridge. These long-lived isotopes, especially berkelium-249 (half-life 320 +/- 3 days), have been effectively utilized in chemical research and as targets for discovery and determination of properties of superheavy (transactinide) isotopes.

Pierce had the vision to commission and support DOE's sponsorship of a National Academy of Sciences workshop on transplutonium elements in February–March 1983. Gerhard Friedlander, an esteemed nuclear chemist, served as workshop chair and Henry Taube was session chair on inorganic chemistry. Friedlander and Taube represented scientific experts who were non-partisan; they had neither a reputation in nor a vested interest in heavy-element chemistry. This workshop revitalized the study of transplutonium chemistry and physics in the United States for the next two decades.

Pierce guided the American Chemical Society Division of Nuclear Chemistry and Technology for years as division councilor, continuing today in the less formal role of division councilor emeritus. Even after retirement, Pierce continued to be active, authoring the influential 1998 report, "The Education and Training of Isotope Experts," which was delivered to the subcommittee on energy and science of the Committee on Science of the U.S. House of Representatives.

Events near the end of the Cold War era—the Chernobyl nuclear reactor accident in 1986, the fall of the Berlin Wall in 1989, and the breakup of the Soviet Union in the early 1990s—signaled a shift in actinide research from nuclear weapons production and nuclear reactor technology to nuclear stockpile stewardship and mitigation of the environmental effects of the Cold War nuclear legacy. The change in world vision and public policy led the Clinton administration in 1994 to eliminate reactor and reprocessing research and much other nuclear research and development from the DOE portfolio.

Global dialogues

Thanks to the Atoms for Peace initiative, which lifted secrecy of all nuclear literature, international actinide science meetings were initiated with the first International Conference on Plutonium in 1957 in Chicago, Illinois. This was followed by Plutonium 1960 in Grenoble, France, which centered on the properties of this most unusual metal; Plutonium 1965 in London, England, which expanded the scope to ceramic materials and their behavior under irradiation; Plutonium 1970 and Other Actinides in Santa Fe, New Mexico;

Plutonium and Other Actinides and the Transplutonium Element Symposium in Baden-Baden, Germany in 1975; and the International Conference on the Electronic Structure of the Actinides in 1978 in Grenoble. The scope of these meetings was enhanced by the maturity of actinide research in the European Community and Japan, but somewhat inhibited by continuing Cold War ideological barriers. These barriers began to be lifted in the 1980s.

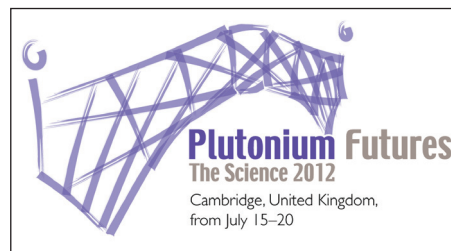
An international quadrennial series of heavy-element research conferences picked up where the earlier plutonium conferences left off; the first was Actinides 1981 in Asilomar, California. Subsequent conferences in the series included Actinides 1985 in Aix en Provence, France; Actinides 1989 in Tashkent, U.S.S.R.; Actinides 1993 in Santa Fe; Actinides 1997 in Baden-Baden; Actinides 2001 in Hayama, Japan; Actinides 2005 in Manchester, United Kingdom; and Actinides 2009 in San Francisco, California.

A number of heavy-element-related conferences that were initiated continue today. The Scientific Basis for Nuclear Waste Management symposia began in 1978 as part of Materials Research Society conferences; the most recent was Scientific Basis for Nuclear Waste Management XXXIV, held in April 2010 in San Francisco.

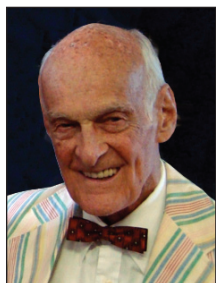
The International Conferences on the Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere—known as the Migration conferences—have been held biennially since 1987. They provide an international forum on research essential for understanding long-term nuclear waste disposal. The first Migration conference was held in Munich, Germany. Besides Germany, other venues have included the United States, Spain, France, Japan, Austria, Korea, and France. The 13th international conference is scheduled for September 2011 in Beijing, China.

A series of conferences entitled Plutonium Futures—The Science was initiated by Los Alamos in 1997 to renew the tradition of open discussions of fundamental properties of plutonium and related elements. The vision and strong support of Los Alamos director Siegfried Hecker and associate director Paul Cunningham were key to establishing this conference series. Venues included Santa Fe (1997 and 2000) and Albuquerque (2003), New Mexico; Asilomar, California (2006); Dijon, France (2008); and Keystone, Colorado, where the most recent conference was held in September 2010. Plutonium Futures 2012 is planned for Cambridge, United Kingdom.

The Office of Basic Energy Sciences sponsored ten Basic Research Needs workshops from 2002 through 2007. One of these was the 2006 workshop on Advanced Nuclear Energy Systems, chaired by James Roberto of Oak Ridge National Laboratory and Tomas Diaz de la Rubia of Lawrence Livermore National Laboratory. Most of the U.S. actinide science community participated in this workshop, which identified new and refocused challenges in actinide coordination chemistry, separations, spectroscopy, materials science, and long-term behavior of actinide waste forms.



The Atoms for Peace initiative opened an avenue for global international dialogues beginning with the International Conference on Plutonium, held in Chicago, Illinois, in 1957. Since then, researchers have traveled literally around the globe to attend conferences in Austria, France, Germany, Japan, Korea, Spain, the United Kingdom, and the U.S.S.R., as well as the United States. Beijing, China, will be the venue for the 13th Migration conference this September, and Cambridge, United Kingdom, will be the site of Plutonium Futures—The Science 2012.



Elliot Pierce



Patricia Dehmer

While serving as director of the Office of Chemical Sciences, Elliot Pierce, encouraged the Department of Energy's sponsorship of a 1983 National Academy of Sciences workshop on transplutonium elements that revitalized the study of transplutonium chemistry and physics in the United States for the next two decades.

During her tenure as director of the Office of Basic Energy Sciences, Patricia Dehmer led an effort to relate fundamental research in energy to real-world problems. She also oversaw the planning of five Department of Energy nanocenters that serve as premier user centers for interdisciplinary research.

The number of university research grants awarded by the Office of Basic Energy Sciences increased dramatically under Norman Edelstein's guidance. He was also instrumental in initiating a series of heavy-element contractor meetings, which continue today.



Norman Edelstein

Actinide research today

The BES Heavy Element Chemistry program remains a key source of federal support in the United States for fundamental research on the chemistry of the actinides and their fission products. Within the Office of Science, Patricia Dehmer served as the director of BES from 1995 to 2007. Under her leadership, the BES budget more than doubled to \$1.2 billion annually.

Dehmer's DOE biography credits her with building "a world-leading portfolio of work in condensed matter and materials physics, chemistry, and biosciences. A five-year effort to relate fundamental research in these disciplines to real-world problems in energy—including problems in fossil energy and carbon dioxide sequestration, nuclear energy, renewable energy, energy efficiency, energy transmission and storage, and the mitigation of environmental impacts of energy use—facilitated greater integration of basic and applied research across DOE."

During her tenure at BES, Dehmer was responsible for the planning, design, construction, and operational support of large research facilities, including the Spallation Neutron Source at Oak Ridge, built by a partnership of six DOE laboratories; the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory (formerly known as the Stanford Linear Accelerator Center); and five nanocenters.

DOE considers the five nanocenters to be the "premier user centers for interdisciplinary research at the nanoscale, serving as the basis for a national program that encompasses new science, new tools, and new computer capabilities." The centers are located at six national laboratories: Oak Ridge (the Center for Nanophase Materials Sciences), Lawrence Berkeley (the Molecular Foundry), Sandia and Los Alamos (the Center for Integrated Nanotechnologies), Brookhaven (the Center for Functional Nanomaterials), and Argonne (the Center for Nanoscale Materials).

Dehmer, now deputy director for science programs in the DOE Office of Science, currently has oversight for the office's six science programs: basic energy sciences, biological and environmental research, fusion energy sciences, advanced scientific computing research, high-energy physics, and nuclear physics. The Office of Science supports research at some 300 colleges and universities nationwide, as well as at the DOE laboratories and private institutions.

In 2000 acting BES program manager Norman Edelstein initiated BES heavy-element chemistry contractor meetings. These meetings continue biennially as information-exchange forums in the spirit of the Gordon Research Conferences. (The Gordon Conferences are international forums in which investigators from around the world can discuss their latest research and future challenges in an informal setting. The first Gordon Conference was held in 1931 in a Baltimore classroom. Since then, more than 5,500 weeklong conferences have been held, attracting more than 600,000 attendees.)

Under Edelstein's guidance, the number of grants awarded to university actinide researchers increased dramatically. The grants' scope ranged from organoactinide chemistry to theoretical studies of actinide dioxides and metal surfaces. Collaborative research contracts were awarded to actinide researchers at six institutions under the Russian Academy of Sciences after proposals were solicited and peer reviewed. These research activities were carried out with the guidance and active collaboration of U.S. actinide scientists at national laboratories and universities. Transport of actinides on colloids, aggregates, and nanoparticles was an innovative aspect of these binational collaborations carried out between 2001 and 2008.

New in 2009 for BES were Energy Frontier Research Centers and Single-Investigator and Small-Group Research projects. Awards under these initiatives led to a 40-percent increase in the heavy-element chemistry budget, from about \$10 million annually to about \$14 million annually.

Paralleling the trend of many other research areas of physical science, theoretical advances in heavy-element chemistry now claim a partnership role with experimental advances. Advances in density functional theory have made it possible to model multi-atom systems that have significant relativistic and spin-orbit effects. Modeling of actinide atoms and ions requires theoretical treatment of these effects; such modeling is now successful for actinide metals, actinides on surfaces, actinides in gaseous species, and even in actinide ions that undergo oxidation-reduction reactions in aqueous solution.

Heavy-element theorists can suggest to experimentalists synthesis of new species and remeasurement of some data that may be incorrect and should be reconsidered. Experimentalists now partner with theoreticians by confirming the insights and quantitative results now achievable by theoretical methods as well as by measuring properties that calibrate theoretical calculations.

A final thought

"Science has its cathedrals, built by the efforts of a few architects and of many workers," says Coffey in *Cathedrals of Science*. While the "cathedral" of actinide science in the United States is no longer in a nascent stage, it is still developing and remains in need of the continued support of the government and its citizens. The role of the scientific leaders whose managerial vision fostered the growth of actinide science in the United States cannot be overemphasized and four "architects" of the U.S. heavy-element chemistry program deserve another mention:

Vannevar Bush, who led the National Defense Research Committee, the Uranium Committee, and the Office of Scientific Research and Development, and who guided the basic research tradition of peer-reviewed proposals in the National Science Foundation and the DOE Office of Science.

Glenn T. Seaborg, for his leadership in transuranium element science at Berkeley, at the Metallurgical Laboratory, and as Atomic Energy Commission chairman, as well as for his visionary championing of basic research and science education.





Elliot Pierce, for commissioning the 1983 workshop on transplutonium elements and for maintaining sponsorship of transplutonium element production, separation, and research in that decade.

Patricia Dehmer, for initiating the Basic Research Needs Workshop for U.S. Energy Security, which led to appreciation for the need for continued basic research in heavy-element science as well as funding enhancements in the first decade of the twenty-first century.

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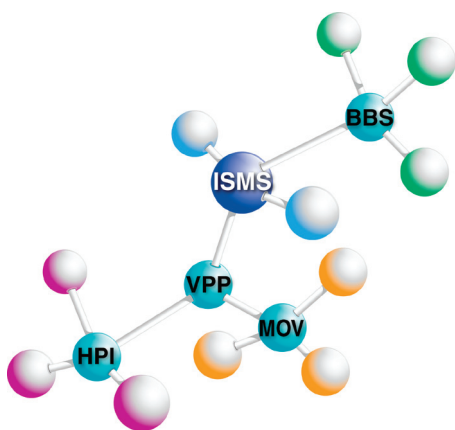
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Patricia Worthington, director, DOE Office of Health and Safety

ATOMICS: A REACTION TO SAFETY GOES LABWIDE

Ten years ago, Los Alamos management implemented a behavior-based safety (BBS) program called ATOMICS at the Chemistry and Metallurgy Research Building at TA-3 and the Plutonium Facility at TA-55. The mission of ATOMICS—an acronym for Allowing Timely Observations Measures Increased Commitment to Safety—is to improve the health and safety of the Laboratory’s workforce by engaging employees in working together to identify and eliminate at-risk behaviors and conditions through ongoing safety observations.

The program has experienced significant growth since its inception, due largely to enhancements made to the process by Mike Mallory, principal associate director for Business and Operations and formerly associate director for Stockpile Manufacturing and Support, and Carl Beard, associate director for Plutonium Science and Manufacturing. Both managers came to Los Alamos from BWXT Pantex in Texas, where a behavior-based observation program is one of several safety tools used at the facility with great success.

In 2007, the ATOMICS process at TA-55 was reintroduced to Stockpile Manufacturing and Support employees, and in the following two years, workers began an aggressive effort to perform observations regularly at TA-55 and at the TA-22 Explosives Detonator Facility. (See *ARQ* 4th Quarter, 2007.) The Plutonium Manufacturing and Technology Division saw a 64-percent reduction in the number of total reportable cases from fiscal year 2008 to fiscal year 2009.

Initially, workers were trained as observers to identify safe and at-risk worker behaviors. The scope of observations has been expanded to include workplace conditions. The enhanced ATOMICS program also puts additional emphasis on data analysis, using leading indicators to identify root causes of injuries and to track safe work practices. As observations are made, at-risk behaviors or conditions that can’t be immediately resolved at a team or group level are entered into the Laboratory’s ATOMICS database and tracked until they are resolved.

“During the ISM Champions Workshop, I had the opportunity to follow up on Los Alamos’ ATOMICS program, and I was encouraged by its progress and maturity. I found the blends of behavioral- and human-performance-based observations and management ownership of the program to be important elements of an effective approach for reinforcing safe behaviors and eliminating at-risk behaviors and conditions for employees and operations.” —Patricia Worthington

The Environment, Safety, and Health Integration Office (ESS-IO) manages the Lab’s BBS ATOMICS program, and facilitators Jim Kleinsteuber and Maryrose Montalvo work with other subject-matter experts to ensure



integration of sustainable safety practices into every aspect of the Laboratory's mission. The program incorporates insights from Human Performance Improvement (HPI) concepts, the Management Observation and Verification (MOV) process, and Voluntary Protection Program (VPP) criteria.

Recently, the ATOMICS Orientation course received Training Staff Qualification (TSQ) in accordance with a DOE order affecting training requirements for nuclear facilities. "The TSQ credential indicates that the course and the instructors will implement training that is effective and DOE compliant, and that meets the mission support needs of the organization," said training specialist Brenda Fernandez. "This credential is the standard we strive to achieve for all classroom instructors throughout the Laboratory."

"Formality of operations at the TA-55 Plutonium Facility is among the most rigorous in the entire Laboratory. Consequently, the facility has been on the leading edge of implementing a behavior-based safety program. The basic approach is two-fold. First, to raise worker participation in our local ATOMICS program to achieve effective contact rates between employees, which will ultimately result in decreased injury rates. Second, to aggressively identify leading indicators (as opposed to lagging indicators such as total reportable cases and days away, restricted or transferred) and thus improve systems that if not addressed could result in more consequential events. The outcome has been that our employees now view safety as an integral part of their normal workday, and they even take this approach home to share with their families and friends." —Steve Schreiber

Today, ATOMICS is an institutional safety tool used by technicians, engineers, and support personnel all over the Laboratory. Los Alamos organizations have recognized the safety successes within the Plutonium Science and Manufacturing Directorate and have requested support to strengthen their own safety programs. Thirteen organizations—ranging from the Director's Office to the Weapons Engineering Directorate—have implemented the program. More than 2300 observers have been trained to date, 400 of them in the Plutonium Science and Manufacturing Directorate alone. Approximately ninety Environmental Programs Directorate workers were trained last fall, as were all fourteen members of the latest Radiological Control Technician Program, many of whom were deployed to TA-55 early this year.

In June of last year, the Security and Safeguards Division asked for assistance to begin a division-wide ATOMICS implementation, in addition to a separate implementation for the Laboratory's Protective Force, SOC Los Alamos. The undertaking with SOC is unique because SOC is a contractor with different sets of contractual agreements with LANS and DOE. Kleinstueber and Montalvo worked closely with Valerie Miranda, director of Compliance and Support



Steve Schreiber, division leader, Nuclear Component Operations



Recently certified radiological control technicians (RCTs) perform routine monitoring activities while veteran RCT Harold Chacon (background) completes an ATOMICS observation.





Tim George (left), deputy director, Plutonium Science and Manufacturing, and Jim Kleinsteuber, ATOMICS facilitator



Crestina Vigil (right), Nuclear Process Infrastructure Group, and Maryrose Montalvo, ATOMICS facilitator

More information on ATOMICS is available on the web at <http://int.lanl.gov/orgs/adeshq/atomics/index.shtml>.

SOC-LA, and with the Laboratory's Intellectual Property and Technology Transfer Office to obtain a copy of the existing ATOMICS database for SOC. This effort will allow SOC employees to enter and review observations at any time, using data sheets from various Laboratory sites.

Ten years ago, Tim George, who was then the leader of Nuclear Materials Technology (NMT) Division, and ATOMICS facilitator Jim Kleinsteuber reviewed a mockup of a cause-and-effect relationship between a heightened number of ATOMICS observations and NMT's total reportable cases (TRC) and days away, restricted or transferred (DART) rates. ATOMICS data tracked over eight years indicated that as worker observations increased, TRC and DART rates decreased proportionally. In contrast, as worker observations decreased, TRC and DART rates increased.

Long-term improvement in behavior-based safety can be achieved by identifying and eliminating organizational weaknesses and building robust defenses within work processes. This is an area in which the Laboratory's Worker Safety and Security Teams (WSSTs) are beginning to make headway, according to Kleinsteuber. Because the WSSTs are typically composed of a cross section of different types of workers (technical and administrative), a variety of organizational perspectives can be incorporated into the ATOMICS process.

Los Alamos' 2011 institutional performance metrics include an objective to continue to implement behavior-based safety processes across the Laboratory. "Participating in safety observations is part of our everyday work operations and reflects our commitment to take care of each other while strengthening LANL's safety posture," said Deputy Laboratory Director Ike Richardson. "If we positively reinforce safe behaviors, test the effectiveness of our actions, document the results, and refuse to be satisfied with our current performance, we will be the best organization in the DOE complex and one of the best anywhere."

"Nearly ten years ago, as an undergraduate student, I had my first experience with ATOMICS. The process was fairly new, and one of my first tasks was to enter data from handwritten cards into the database. Back then I did not always understand the scenarios as described by the workers because I was at a different site. Now, as a resident technician at TA-55, not only do I understand the scenarios, I am part of them on a daily basis. I see firsthand how performing and recording observations build a sound safety culture within the workforce. Today, I still review observations submitted by my peers and present trends, highlights, and significant comments during group meetings." —Crestina Vigil

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