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Fourth Quarter 2019

The Manhattan Project

Scientific and Historical Impacts



Foreword

The Manhattan Project marked a defining moment in human history—the onset of the Nuclear Age. Los Alamos National Laboratory (LANL) played a pivotal role as a key scientific institution in this military undertaking which developed the first technological use of uranium and plutonium in atomic bombs. The effort brought swift closure to World War II and ushered in the Nuclear Age, initiating global nuclear security, international policies of nonproliferation, nuclear science and technology, and actinide chemistry, metallurgy, and materials science. The Manhattan Project also helped create the mold for international scientific institutions to address the greatest technical challenges of the day.

The Manhattan Project: Student Symposium was held on July 17, 2019, at LANL. It was jointly organized by Dr. A. Balatsky and the leadership of the Institute for Materials Science and National Security Education Center to discuss the long-standing impact the Manhattan Project and its scientific and technical staff have had on international science, politics, and global security. The idea to have a student symposium focused on the Manhattan Project grew out of conversations with colleagues and students—the prevailing opinion is that there is a growing interest in history, science, politics, and security implications of this event. We thought it would be beneficial to enable continued dialog and discussion with younger generations, providing them the opportunity to formulate their own opinions.

We assembled an excellent team of experts who spoke with authority on the topics: Senior Los Alamos Historian Dr. Alan Carr presented the history of the Manhattan Project (p2); former Los Alamos Director Prof. Siegfried S. Hecker discussed the role of plutonium as a key material (p14); Los Alamos Fellows Dr. Mark B. Chadwick, Dr. David L. Clark (p20), and Dr. James L. Smith (p28) discussed the associated interdisciplinary science: metallurgy, materials science, chemistry, waste disposal, and particle physics. Finally, Dr. Galya Balatsky and Dr. Parrish Staples outlined the challenges of nonproliferation in the post-Cold War era (p32). We also hosted a roundtable discussion moderated by Dr. Joseph C. Martz at the end of the day, with the experts fielding questions from the student body.

This special edition of Actinide Research Quarterly (ARQ) is dedicated to capturing the Manhattan Project: Student Symposium event and its content. The Manhattan Project is an example of “living history” where multiple generations continue to learn and evaluate the full magnitude of its history, science, technology, deterrence, and security implications at different times. We hope that this issue helps illuminate some of these subjects for the readers of ARQ. The editors also wish to thank the speakers for assembling this information and delivering it in such an accessible format for future generations of scientists, engineers, researchers, and technical staff.

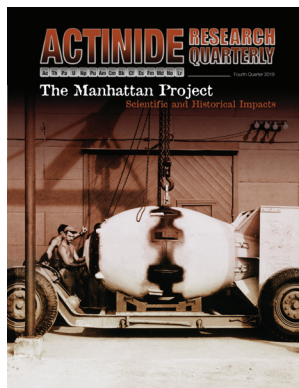
Alexander Balatsky *University of Connecticut*

Filip Ronning, Director *Institute for Materials Science*

Franz J. Freibert, Director *G.T. Seaborg Institute*



About the cover: This recently declassified image shows the assembly of the Fat Man device at Tinian Island in the South Pacific prior to loading onto the Boeing B-29 Superfortress Bockscar, August, 1945. This implosion bomb, the first of its kind to use plutonium fuel, was developed by the Manhattan Project and quickly superseded its predecessor, the uranium-based gun-type design Little Boy, due to its more efficient use of fissile material. This feat was particularly remarkable because elemental plutonium was only identified and isolated a few years earlier in 1940.



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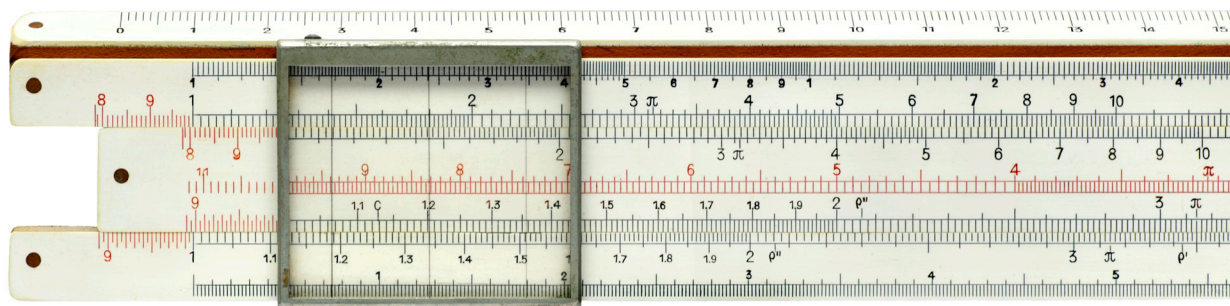
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Alan B. Carr

Alan B. Carr currently serves as a Program Manager and the Senior Historian for Los Alamos National Laboratory. During his 16 years at the Lab, Alan has produced several publications pertaining to the Manhattan Project, nuclear weapons testing, and the Laboratory's development during the Cold War years. He has lectured for numerous professional organizations and been featured as a guest on many local, national, and international radio and television programs. Before coming to Los Alamos, Alan completed his graduate studies at Texas Tech University in Lubbock.

Manhattan: The View from Los Alamos of History's Most Secret Project

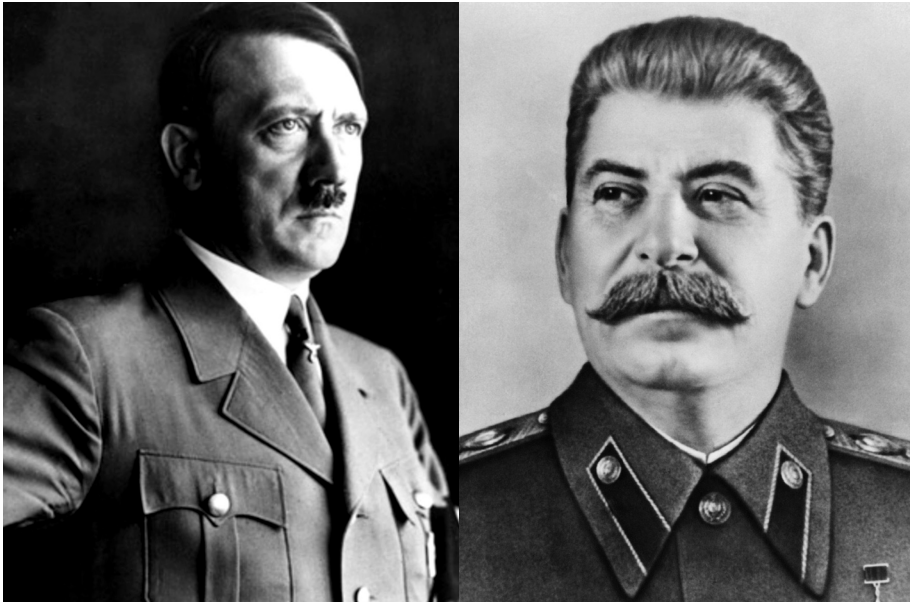
Alan Carr

Los Alamos National Laboratory, Los Alamos, New Mexico

World War II, history's deadliest conflict, claimed between 60 and 80 million lives worldwide. Months before the war started, scientists first produced nuclear fission in Nazi Germany. Although scientists recognized that this new process could be harnessed in the form of a weapon, it took years for policymakers in the United States to acknowledge that nuclear weapons were both a feasible and transformative new technology that was within reach. In response, the Manhattan Project came into existence in the summer of 1942 to build reliable nuclear weapons as quickly as possible. At the center of the project was a relatively small facility in northern New Mexico tasked with designing, building, testing, and helping deliver America's nuclear weapons in combat. During the war, this secret laboratory was only known by its codenames: Project Y, Site Y, and The Zia Project. Today, it is recognized around the world as Los Alamos National Laboratory; this paper presents the view from Los Alamos of history's most secret project.

On September 1, 1939 the German Army invaded Poland from the west to start World War II. On September 17, the Soviet Union's Red Army invaded Poland from the east. A week earlier, the two nations had signed a non-aggression pact including a secret protocol which divided Poland between the two. France and Britain immediately declared war on Hitler's Germany, but reluctantly maintained neutrality with Stalin's Soviet Union. In the coming months, Stalin invaded Finland and forcefully annexed Latvia, Lithuania, Estonia, and large tracts of Romania. Hitler meanwhile successfully invaded Denmark, Holland, Belgium, Luxembourg, and France. After failing to force a British surrender during the Battle of Britain, Hitler turned eastward believing Britain no longer posed a strategic threat. The morning of June 22, 1941, Hitler broke the non-aggression pact with Stalin and invaded the Soviet Union: known as Operation Barbarossa, it would prove to be history's largest military campaign. Despite suffering millions of casualties in the opening months of Barbarossa, the Soviet Union was able to survive. Nonetheless, the German Army advanced to the gates of Moscow by December 1941 where it was finally halted by exhaustion, freezing temperatures, and a ferocious Soviet counterattack.

As the Soviets defended their capital, the Imperial Japanese Navy invaded the Philippines and launched a surprise attack against the United States Pacific Fleet at Pearl Harbor. Months earlier, in response to Japan's brutal occupation of Southeast Asia, the US imposed significant sanctions on Japan, including the embargo of resources crucial to the war effort such as crude oil and scrap metal. Rather than curbing aggression, the Japanese conceived the plan for the attack on Pearl Harbor. It was hoped the strike would yield a quick and decisive victory over the United States, which was still reeling from the Great Depression. But on the contrary, this attack would rejuvenate the US economy and, after years of fighting, result in the complete annihilation of Imperial Japan.



Hitler and Stalin start World War II: they secretly divided Poland in August 1939. Germany invaded Poland on September 1; the Soviet Union invaded on September 17.



Months before the war started, German scientists produced nuclear fission. At that time, no country was better poised to turn this process into a bomb than Nazi Germany. In addition to having some of the world's greatest scientists, Germany also had a tradition of excellent engineering, significant manufacturing capabilities, and direct access to uranium ore. But, as was the case in the United States, the German

War breaks out in the Pacific: on December 7, 1941, the Japanese attacked the United States Pacific Fleet at Pearl Harbor. The US and Britain immediately declared war on Japan. On December 11, Germany and Italy declared war on the US.



government did not recognize the transformative nature of nuclear weapons. It is well-known that Albert Einstein, at the urging of Hungarian-born physicist Leo Szilard, wrote a letter to President Roosevelt warning him of Germany's nuclear potential. However, Einstein and Szilard described the potential weapon as such: "A single bomb of this type, carried by boat and exploded in a port, might very well destroy the whole port together with some of the surrounding territory." More than 20 years earlier, an ammunition ship exploded in the port of Halifax, Canada, destroying the whole port together with some of the surrounding territory. Because Einstein and Szilard had described an accident, not a transformative weapon, American research in the years that followed would focus on producing reactors for electricity rather than nuclear weapons.

Weeks before the Battle of France began, two Axis-born physicists, Otto Frisch (Austria) and Rudolf Peierls (Germany) working at the University of Birmingham, penned a letter to the British government describing a practical design for a nuclear weapon that could be delivered by air. Willing to pursue a decisive weapon that might turn the tide of the war, the government sponsored a feasibility study to further explore the Frisch-Peierls concept. Days after the German invasion of the Soviet Union the study, known as the MAUD Committee Report, was completed. It confirmed the Frisch-Peierls proposal: transformative, air-deliverable nuclear weapons were most likely only a few years away.

Britain now had an ally of convenience in the Soviet Union courtesy of the German invasion, however most agreed the Soviet Union would wither quickly amidst the German onslaught. Britain had hoped to bring the United States into the war since its onset, but to no avail. As the Germans pushed deep into the Soviet Union, the British Government sent a special mission to the US armed with a copy of the MAUD Committee Report. British leaders hoped to show the Americans that nuclear weapons were at-hand, raising the inevitable question: how close might the Germans be to perfecting a nuclear bomb? Unfortunately, for many months the MAUD Committee Report was ignored in the US, until senior scientific advisors recognized its significance in the days leading-up to Pearl Harbor. Just as officials began thinking about a fission weapon in more serious terms, the Japanese attack and the subsequent German declaration of war on the United States created a new set of priorities. Still, as the early months of 1942 passed, the question of a German nuclear monopoly remained.

Finally, in the summer of 1942, the Manhattan Project was born. The small committees and offices that had overseen the government's nuclear research were largely replaced by the Army Corps of Engineers. The Army established the initial headquarters in Manhattan, hence the project's iconic name. Colonel Leslie Groves, a highly-educated and experienced engineer who had built the Pentagon in approximately 18 months, was selected to lead the project. In addition to a promotion to General, Groves was given the highest priority for labor and war materials, as well as an unlimited budget. He was also introduced to a man many considered to be America's leading theoretical physicist: J. Robert Oppenheimer.

At the peak of the Manhattan Project, General Groves employed nearly 130,000 employees simultaneously at sites all over the country. The three main installations included Oak Ridge, Tennessee—where uranium would be enriched—and Hanford, Washington, which would produce plutonium. Oppenheimer, who had thoroughly impressed Groves, was selected to lead the third site: the project's weapons design laboratory. As the Germans besieged Stalingrad, Manhattan Project officials searched for a suitable location: the laboratory had to be remote, far inland, near a rail line, and the land would have to be easy to acquire. New Mexico, a place Oppenheimer knew well, seemed ideal. An area known as Los Alamos was selected late in 1942 and its inhabitants, the students and staff of a school for boys and several local homesteaders, were promptly evicted. In early 1943, the Laboratory and a small, adjoining community were constructed. In April the first major technical conference was held to baseline the staff's knowledge of nuclear science. Later that month, the University of California signed a contract to operate the Laboratory on behalf of the Army, lending its illustrious name to the Manhattan Project in the national interest during a time of war. This gave Oppenheimer a powerful recruiting tool, in that he could offer prospective staff members employment with the University.

Back on December 2, 1942, Italian Nobel Laureate Enrico Fermi's team at the University of Chicago initiated the world's first controlled nuclear chain reaction. Arguably history's most significant individual scientific experiment, the reaction confirmed nuclear weapons were possible: if one can produce a controlled chain reaction, one can produce an uncontrolled chain reaction (i.e., a bomb). Encouraged by Fermi's success at Chicago, work progressed quickly at Los Alamos throughout 1943. The main bomb design, codenamed Thin Man, was a gun-assembled plutonium device. Another gun-assembled weapon, called Little Boy, was developed concurrently with Thin Man. In a gun-assembled weapon, a fissile projectile is fired at a fissile target to achieve supercriticality. Thin Man was the preferred design because it used plutonium, a more energetic material that could be produced far more easily than enriched uranium. Unfortunately, in the spring of 1944, Project Y suffered a major setback when future Nobel Laureate Emilio Segrè discovered plutonium would not work in a gun-assembled device: an overabundance of neutrons would cause the device to pre-initiate before it fully assembled, resulting in a non-nuclear fizzle. In response, Oppenheimer reorganized the Laboratory to construct an imploding plutonium bomb dubbed Fat Man. In Fat Man, thousands of pounds of high explosives would be used to compress a sphere of plutonium to achieve supercriticality. If Fat Man worked, the payoff would be immense: the weapon would be very efficient and, unlike Little Boy, it could be rapidly reproduced. But it was late in the war, no one knew how close the Germans might be to producing a nuclear bomb, Fat Man was relatively complicated and it relied entirely on high explosives; a material designed to expand, not implode. As scientists at the Laboratory developed methods to assess implosion tests, Oppenheimer directed Kenneth Bainbridge to prepare for a full-scale test of a Fat Man "Gadget."

Manhattan Project Origins

Albert Einstein
Old Grove Rd.
Nassau Point
Peconic, Long Island
August 2nd, 1939

F.D. Roosevelt,
President of the United States,
White House
Washington, D.C.

Sir:

Some recent work by E. Fermi and L. Szilard, which has been communicated to me in manuscript, leads me to expect that the element uranium may be turned into a new and important source of energy in the immediate future. Certain aspects of the situation which has arisen seem to call for watchfulness and, if necessary, quick action on the part of the Administration. I believe therefore that it is my duty to bring to your attention the following facts and recommendations:

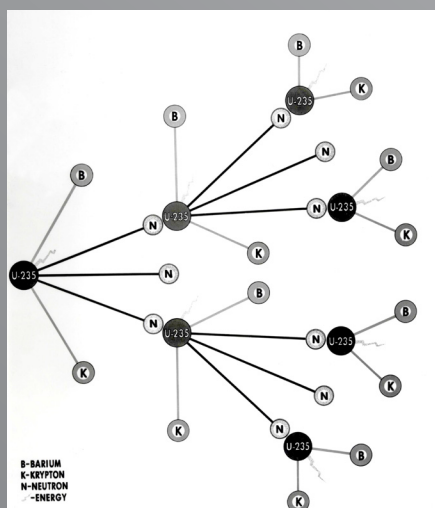
In the course of the last four months it has been made probable - through the work of Joliot in France as well as Fermi and Szilard in America - that it may become possible to set up a nuclear chain reaction in a large mass of uranium, by which vast amounts of power and large quantities of new radium-like elements would be generated. Now it appears almost certain that this could be achieved in the immediate future.

This new phenomenon would also lead to the construction of bombs, and it is conceivable - though much less certain - that extremely powerful bombs of a new type may thus be constructed. A single bomb of this type, carried by boat and exploded in a port, might very well destroy the whole port together with some of the surrounding territory. However, such bombs might very well prove to be too heavy for transportation by air.



The first page of the 1939 letter from Albert Einstein to President Roosevelt warning him of Germany's nuclear potential.

The team of scientists led by Enrico Fermi at the University of Chicago who, on December 2, 1942, achieved controlled nuclear fission for the first time.

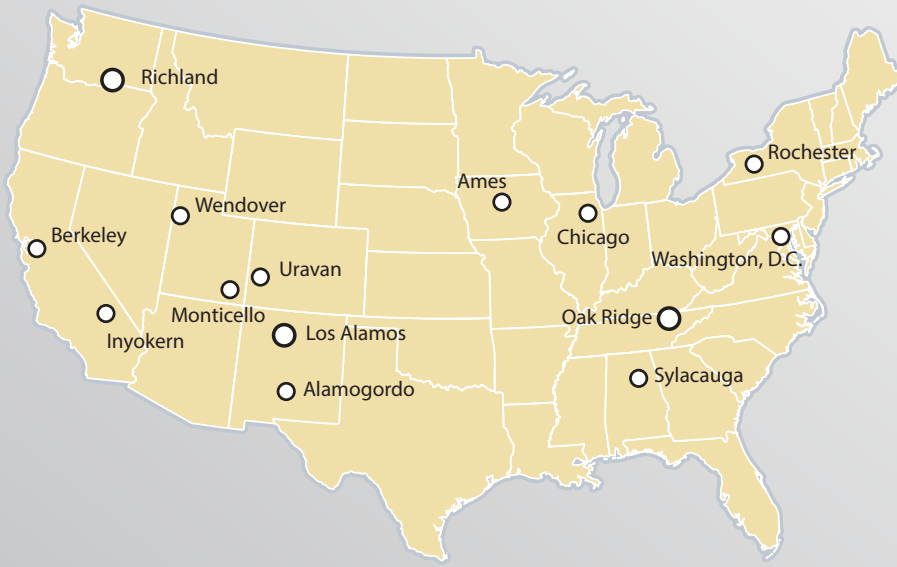


Fission: The splitting of an atomic nucleus resulting in the release of large amounts of energy following a chain reaction.

In Germany, in late 1938, Fritz Strassmann and Otto Hahn produced barium by bombarding uranium with neutrons; Lise Meitner and Otto Frisch identified this process as nuclear fission in early 1939. Scientists immediately realized the potential for an atomic bomb.

A month before Germany invaded Poland, Albert Einstein warned President Roosevelt of fission's potential. In October 1939, Roosevelt responded by establishing the Uranium Advisory Committee. In the summer of 1940, it was absorbed by the National Defense Research Committee; the American project was focused on building a reactor, not a bomb.

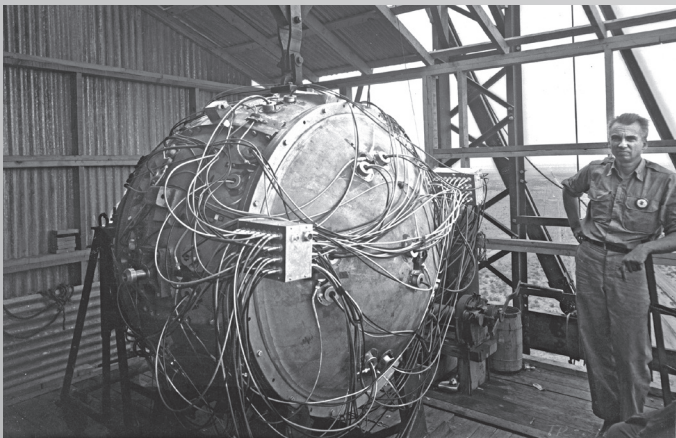
The British MAUD report (July, 1941) predicted an atomic bomb could be completed by late 1943. In fall 1941, the NDRC chairman was given a copy of the report and bomb work accelerated. After the Japanese attack on Pearl Harbor in December 1941 the project was turned over to the Army Corps of Engineers; the Manhattan Engineer District was formally established in August 1942. On December 2, 1942 Enrico Fermi's team at the University of Chicago initiated the world's first controlled nuclear chain reaction.



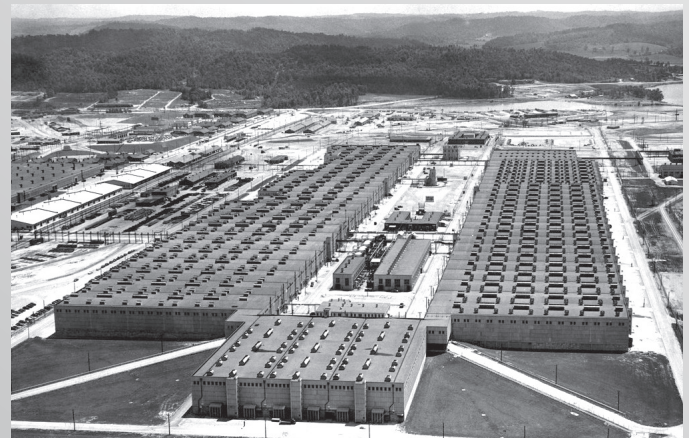
Important Manhattan Project sites in the United States.



General Leslie Groves directed the Manhattan Project.



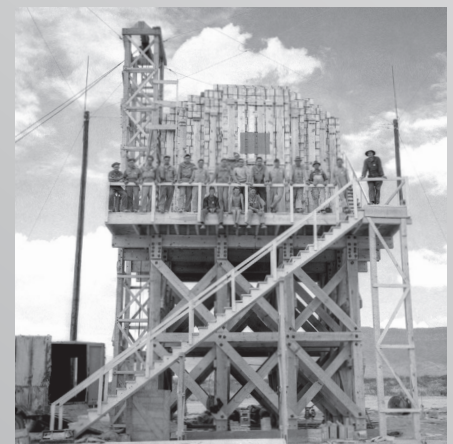
The "Gadget": the prototype plutonium device tested in July 1945.



The Clinton Engineer Works, Oak Ridge, Tennessee, 1943.

In fall 1942, a site in east Tennessee was acquired for a uranium enrichment complex (Oak Ridge). Around the same time, the Hanford Site in Washington was selected for the plutonium production facility. The uranium-235 used in Little Boy was produced at Oak Ridge and the plutonium used in the Trinity device and Fat Man were produced at Hanford.

Groves selected J. Robert Oppenheimer as director of the project's weapons design laboratory, who suggested Los Alamos as a site for the lab. It was established in 1943 as Site Y of the Manhattan Project. The Manhattan Project employed nearly 130,000 workers at its peak with the Los Alamos technical staff comprising between one and two percent of the workforce. The world's first man-made nuclear explosion, the Trinity Test, was successfully detonated July 16, 1945, at the Alamogordo Bombing Range in New Mexico, five weeks after Nazi Germany surrendered.



Crew at the Trinity Test Site, New Mexico.



Top row: The first strike. Hiroshima, a large industrial city with an important army depot, was selected as the target. The gun-assembled uranium weapon, nicknamed "Little Boy," was used on August 6, 1945. The strike completely destroyed five square miles of the city.

Bottom row: The second strike. Kokura, home to one of the largest arsenals in Japan, was selected as the primary target for the second mission. Due to weather, the attack was instead carried out on the secondary target of Nagasaki on August 9, 1945. The implosion-assembled plutonium bomb, "Fat Man," was used. The Japanese yielded to armistice on August 14 and formally surrendered on September 2, 1945.

There would be no full-scale test of Little Boy. Every component of Little Boy was rigorously tested at Los Alamos and, based on those test results, Laboratory scientists were certain the bomb would function in combat. As implosion testing proceeded, the staff grew increasingly confident that Fat Man would also work, but that confidence never translated into certainty. As such, the world's first nuclear weapons test was performed the morning of July 16, 1945. Dubbed Trinity by Oppenheimer, the test produced a yield equivalent to 21,000 tons of TNT and opened a new era in human history: the Nuclear Age.

Nazi Germany collapsed in May 1945, just over two months prior to Trinity. The cost of achieving victory was enormous: during World War II over 300 Americans died in combat, on average, each day. The price was far higher for the Soviet Union, considering 15,000 to 20,000 died on a daily basis due to military action. A vast majority of American and British resources had gone to Europe to help defeat Germany, yet the Allies were able to rout the Japanese in battle after battle with only a small fraction of total resources. Japan had no path to victory, yet continued to fight for several reasons. For instance, in Japan it was considered culturally unthinkable to surrender. Japan lost several battles in World War II, but the country had never lost a war. Although Japanese leaders knew the war was lost, they fought on hoping to extract more favorable terms by inflicting the maximum amount of pain on the Allies. Mounting defeats, conventional bombing, an ever-contracting blockade, and the

大この切なことに書いてあることは最も注
 意して読んで下さい。重大なる秋に
 日本国民諸君は今や重大なる秋に
 直面してしまつたのである。共同宣言
 軍部首脳部の連中が三國共同宣言
 の十ヶ條よりなる寛大なる條項
 を以て此の無益な戦争を止めるが軍
 部は是を無視した。日本に對して宣
 戦米國は今や何人も發明し得なかつ
 亦米國は今や何人も發明し得なかつ
 た。恐ろしい原子爆弾は之を使
 用するに及ばず、原子爆弾は之を使
 一箇口に投下する巨爆弾に匹敵する。

日本國民に告ぐ!!
即刻都市より退避せよ

Thousands of these leaflets were dropped from B-29s over Japan to warn that Hiroshima and other cities might be bombed to total destruction.

public threat of “prompt and utter” destruction after Trinity had not compelled the Japanese to surrender. Before invading the Japanese home island of Kyushu, the Allies would unleash nuclear weapons against Japan, hoping they would bring an abrupt end to the war.

On August 6, the B-29 bomber Enola Gay carried Little Boy into combat against the Japanese city of Hiroshima: the 15 kt blast destroyed five square miles of the city. By the end of November, 1945, 64,500 people—including thousands of Korean forced laborers and approximately ten American prisoners of war—died as a result of the attack. Unfortunately, the Japanese did not surrender. On August 8, the Soviet Union declared war on Japan and invaded Manchuria early the next morning, killing nearly 84,000 Japanese soldiers in the short campaign that ensued. Hours later, the B-29 Bockscar arrived at the city of Kokura with Fat Man armed. However, unable to visually acquire the city below due to cloud cover, the plane left after three bombing runs for the secondary target: Nagasaki. Shortly after 11 AM, Fat Man exploded over the Mitsubishi-Urakami Torpedo Works, the factory that manufactured torpedoes used at Pearl Harbor. Though the blast produced by Fat Man was greater than that of Little Boy (21 kt versus 15 kt), fewer people died because the detonation point was on the outskirts of town. Nonetheless, just over 39,000 people—including thousands of Korean and hundreds of Chinese forced laborers—died before the year was out. A letter penned by future Nobel Laureate Luis Alvarez, with input from fellow Los Alamos scientists Phillip Morrison and Robert Serber, addressed to Japanese physicist Ryokichi Sagane was dropped near Nagasaki, several miles from ground zero. In part it read, “As scientists, we deplore the use to which a beautiful discovery has been put, but we can tell you that unless Japan surrenders at once, this rain of atomic bombs will increase manyfold in fury.” It was a promise the US could have made good on: multiple Fat Man-type units could have been delivered in combat on a monthly basis from that point forward.

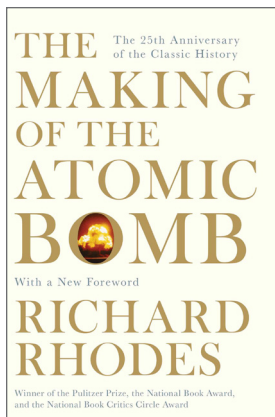
The tragedy of statistics. World War II claimed between 60 and 80 million lives.

	Fatalities
American	418,500 (over 300 died daily)
American Pearl Harbor	2,402
American D-Day	2,499
Soviet	27,000,000 (upper estimate)
Stalingard	2,000,000
Operation Meetinghouse (Tokyo)	100,000
Hiroshima	64,500 (mid-November 1945)
Nagasaki	39,214 (mid-November 1945)
Jewish Holocaust	5,900,000
Chinese	20,000,000 (upper estimate)

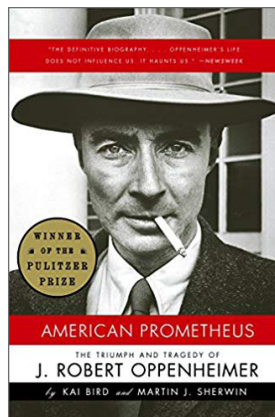
The next day, on August 10, 1945, the Japanese Government informed the Allies it would surrender, provided the agreement “does not comprise any demand which prejudices the prerogatives of His Majesty [the Emperor, Hirohito] as Sovereign Ruler.” In response, Secretary of State James F. Byrnes wrote:

From the moment of surrender the authority of the Emperor and the Japanese Government to rule the state shall be subject to the Supreme Commander of the Allied Powers who will take such steps as he deems proper to effectuate the surrender terms... The ultimate form of government of Japan shall, in accordance with the Potsdam Declaration, be established by the freely expressed will of the Japanese people.

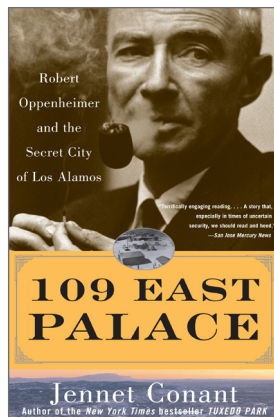
Thus Japan surrendered unconditionally and the armistice went into effect on August 14, but not before tens of millions lay dead among the ruins of a largely destroyed world. There was a celebration back at Los Alamos, but the excitement was tempered by fears that the next world war would feature nuclear weapons. On October 16, General Groves awarded the Laboratory—now publicly known as Los Alamos Scientific Laboratory—the Army-Navy “E” Award for excellence in wartime production. In accepting the award on behalf of Los Alamos, Director Oppenheimer warned: “The people of this world must unite or they will perish. This war that has ravaged so much of the earth, has written these words. The atomic bomb has spelled them out for all men to understand.” Although the unity Oppenheimer called for remains elusive, the threat of nuclear weapons has helped prevent another world war. Today, it remains the mission of Oppenheimer’s Laboratory to develop technologies that will help make the world safer and more sustainable for its inhabitants.



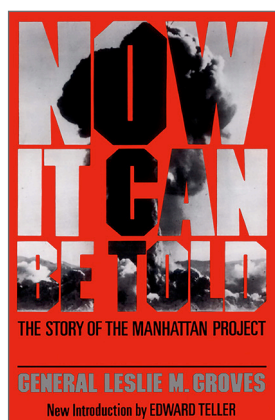
Easily the best all-around history of the entire Manhattan Project. It's a huge book, but written exceptionally well. If you only read one book on the Manhattan Project, make it this one.



The Pulitzer Prize-winning definitive biography of Oppenheimer. I often refer to it as, 'the everything you wanted to know about Oppenheimer and so much more' biography.



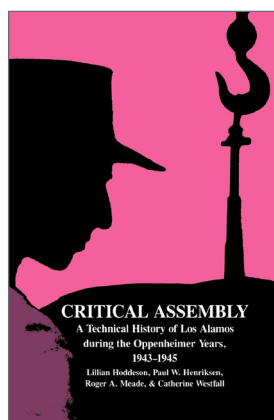
A popular and readable account of Project Y that focuses more on the social aspects of Los Alamos largely from the perspective of Dorothy McKibbin, the secretary who ran the Lab's Santa Fe office.



General Groves' memoir. Like all memoirs, it should be handled with caution! That said, I think it's pretty good.



This book is a bit esoteric, but does give a good—though clinical—account of the safety standards during the Manhattan Project.



An excellent technical history of the wartime years at Los Alamos. The authors had full access to the LANL Archives. It's considered the Lab's official history of the project.

Further reading:

1. "The Making of the Atomic Bomb," Richard Rhodes, Simon & Schuster, 2012.
2. "American Prometheus: The Triumph and Tragedy of J. Robert Oppenheimer," Kai Bird and Martin Sherwin, Vintage Books, 2006.
3. "109 East Palace: Robert Oppenheimer and the Secret City of Los Alamos," Jennet Connant, Simon & Schuster, 2006.
4. "Now it Can Be Told: The Inside Story of the Development of the Atomic Bomb," Leslie Groves, Da Capo Press, 1983.
5. "The Dragon's Tail: Radiation Safety in the Manhattan Project, 1942-1946," Barton Hacker, University of California Press, 1987.
6. "Critical Assembly: A Technical History of Los Alamos during the Oppenheimer Years, 1943-1945," Lillian Hoddeson, et al., Cambridge University Press, 1993.



Siegfried S. Hecker

Dr. Hecker is a Research Professor Emeritus at Stanford University in the Center for International Security and Cooperation. He is a former Director of Los Alamos National Laboratory (1986–1997). Dr. Hecker's current research interests include plutonium science, nuclear weapons policy, nuclear security, and the safe and secure expansion of nuclear energy. Over the past 25 years, he has fostered cooperation with the Russian nuclear laboratories to secure and safeguard the vast stockpile of ex-Soviet fissile materials.

Plutonium: Manhattan Project to Today

Siegfried S. Hecker

Center for International Security and Cooperation, Stanford University, Stanford, California

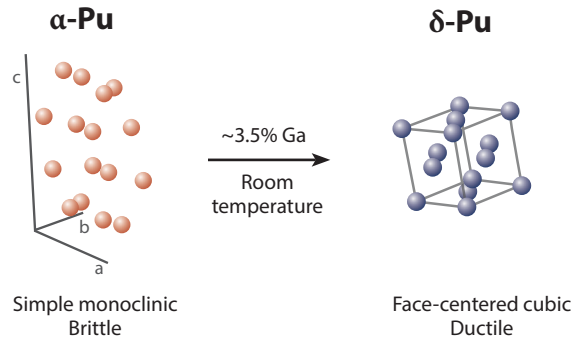
Plutonium symbolizes everything we associate with the nuclear age. It evokes the entire gamut of emotions from good to evil, from hope to despair, and from the salvation of humanity to its utter destruction. No other element bears such a burden. Its discovery in 1941, following the discovery of fission in 1938, unlocked the potential and fear of the nuclear age.

On March 28, 1941, Kennedy, Seaborg, Segre, and Wahl first demonstrated that Pu-239 undergoes slow neutron-induced fission with a cross-section that was approximately 50% greater than that of U-235, releasing millions of times the energy typically released in conventional chemical explosives. This discovery opened the second path to an atomic bomb. The physics of plutonium bombs turned out to be challenging because the gun-assembly technique developed for uranium bombs was too slow, requiring a much more complicated spherical implosion technique. Just as challenging was developing the chemical, metallurgical, and engineering methods to craft plutonium into such spherical assemblies.

Manhattan Project scientists and engineers managed the incredible feat of taking the discovery by Glenn Seaborg and colleagues to expand plutonium production in less than three years from micrograms to the kilograms required for the nuclear bomb that destroyed Nagasaki. What made this feat even more remarkable was that plutonium turned out to be the most complex element in the periodic table.

As element 94, it fits near the middle of the actinide series. It is the 5f electrons that make plutonium extraordinarily complex. With little provocation, the metal changes its density by as much as 25%. It can be as brittle as glass or as malleable as aluminum; it expands when it solidifies—much like water freezing to ice; and its shiny, silvery, freshly-machined surface will tarnish in minutes. It is highly reactive in air and strongly reducing in solution, forming multiple compounds and complexes in the environment and during chemical processing. It transmutes by radioactive decay, causing damage to its crystalline lattice and leaving behind helium, americium, uranium, neptunium, and other impurities. Plutonium damages materials on contact and is therefore difficult to handle, store, or transport. Who would ever dream of making and using such a material? Physicists did during the Manhattan Project—to take advantage of the nuclear properties of Pu-239.

These peculiarities of the newly-created metal were discovered during the frantic years of 1943 to 1945, one surprise after another as the reactors at Oak Ridge and Hanford produced sufficient quantities of plutonium metal to permit characterization. For example, as late as 1944 the measured density of plutonium metal varied from 11 to 22 g/cc because surface reactivity led to severe oxidation and the metal was found to exhibit multiple crystallographic forms, with the room-temperature phase appearing to be brittle as glass. A reproducible density is critical to bomb design and metal ductility was highly desirable for manufacturability.



Cyril Stanley Smith led Manhattan Project efforts to determine the metallurgical properties of plutonium, arguably the most complex metal in the periodic table with six allotropes. Through an alloy screening program it was discovered in 1944 that by adding small amounts of gallium, the ductile δ phase could be attained at room temperature. This allowed the metal to be machined in a manner similar to aluminum, a discovery critical to the development of the atomic bomb.

In a remarkable effort in 1944, Cyril Stanley Smith, the lead metallurgist at Los Alamos, and his colleagues conducted an alloy survey program that led to the production of a face-centered cubic (fcc; δ phase) form of plutonium with a reproducible density of roughly 15.75 g/cc that exhibited ductility akin to that of commercially-pure aluminum, rather than glass. The magic formulation consisted of adding approximately 3.5 atomic percent gallium to plutonium before casting, which led to the retention of the fcc δ -phase at room temperature. It was recognized that this phase is likely in a metastable state, but anticipated requirements were viewed to be months, not years or decades. The “long-time stability” study of the material ran out of time at 44 days because the first devices needed to be fabricated.

The surface of plutonium metal also proved problematic as it oxidized at dramatic rates in certain environments, requiring coating of the plutonium components. The remarkable progress in taming this complex element made by chemists, metallurgists, and engineers during the Manhattan Project is described by one of the pioneers, Edward Hammel, in “Plutonium Metallurgy at Los Alamos, 1943 to 1945.”

Cold War

During the Cold War, the primary interest in plutonium was to provide triggers for thermonuclear weapons for a triad of delivery means (i.e., weapons delivered by bombers and both land-based and sea-launched missiles) that formed the basis of nuclear deterrence. Both the engineering requirements encompassing a large range of temperatures and stresses and the physics requirements for successful detonation became more demanding as the nuclear devices were designed to be smaller and lighter. The manufacturing requirements likewise increased as the US scaled up not only the sophistication of its weapons, but also dramatically increased their number.

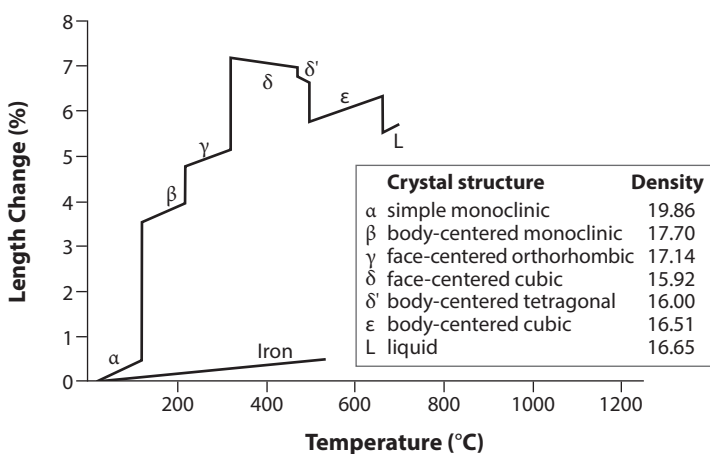
The manufacturing role shifted to the Rocky Flats Plant in 1952. The Los Alamos laboratory continued to play the lead role in the US nuclear complex in plutonium alloy development and property characterization during the Cold War, although significant efforts were mounted at the Lawrence Livermore laboratory along with early work at Argonne and Pacific Northwest laboratories. Moreover, President Eisenhower’s Atoms for Peace initiative launched in December 1953 led to international collaboration on the fundamental properties of plutonium. The first international conferences describing some of this work were the International Conferences on the Peaceful Uses of Atomic Energy held in Geneva in 1955 and 1958.

International conferences dedicated primarily to plutonium were held in 1965, 1970, and 1975, followed later by a variety of such conferences on plutonium and the actinides. The first edition of the plutonium handbook, “Plutonium Handbook: A Guide to the Technology,” was published in 1967. David Clark has led the effort at Los Alamos to publish a seven-volume update, which was released in November.

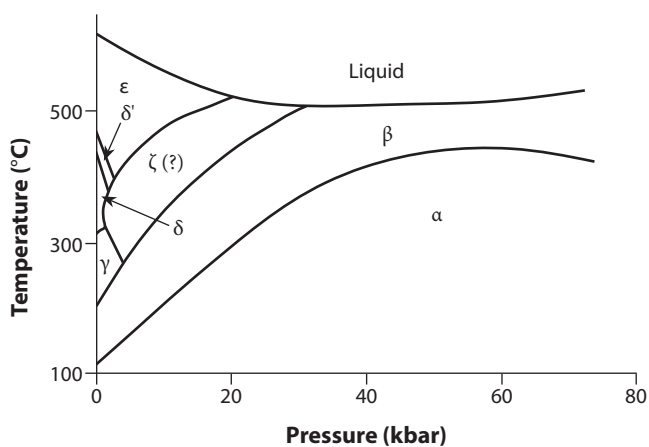
Unusual properties of plutonium

- Unique low-symmetry crystal structures
- Six allotropic phases (seventh under pressure)
- Fcc phase is least dense and highly elastically anisotropic
- Dramatic volume changes
- Extreme sensitivity to alloying
- Low melting point
- Low cohesive energy
- Anomalies in low-temperature transport properties
- Volume decrease upon melting
- Large specific heats
- Dramatic variation in mechanical properties
- Highly unusual properties of the liquid phase
- Very high self-diffusion in bcc epsilon phase
- Great affinity for oxygen and hydrogen
- Very large thermal expansion coefficients
- Negative thermal expansion in fcc phase
- Self-irradiation damage due to radioactive decay

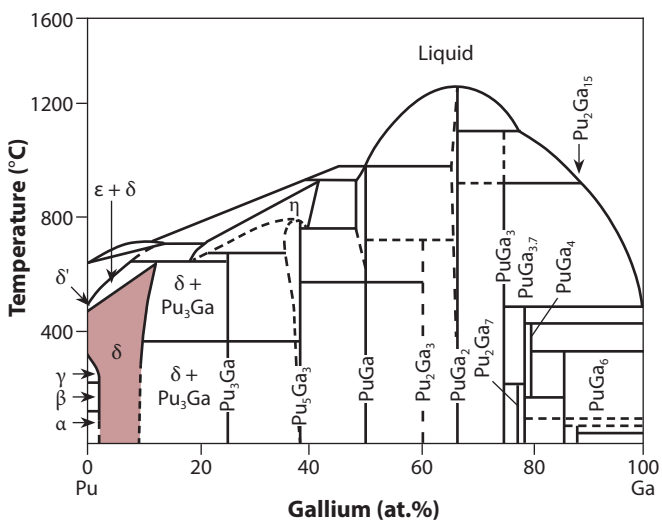
The instability of plutonium demonstrated in four graphs:



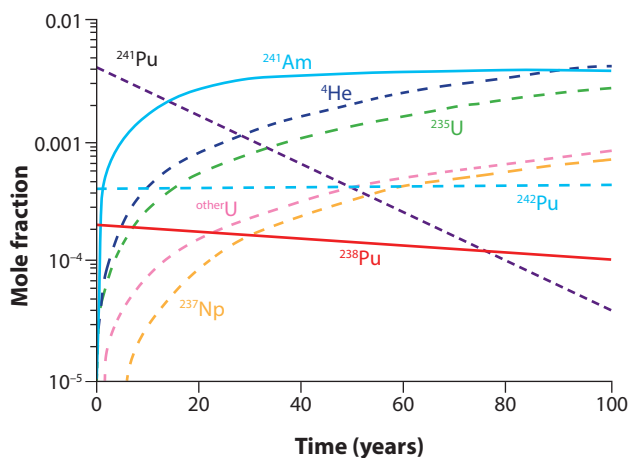
(a) The remarkable effect of temperature on elemental Pu, which displays multiple allotropes and significant volume changes; Fe shown as an example of typical metal behavior.



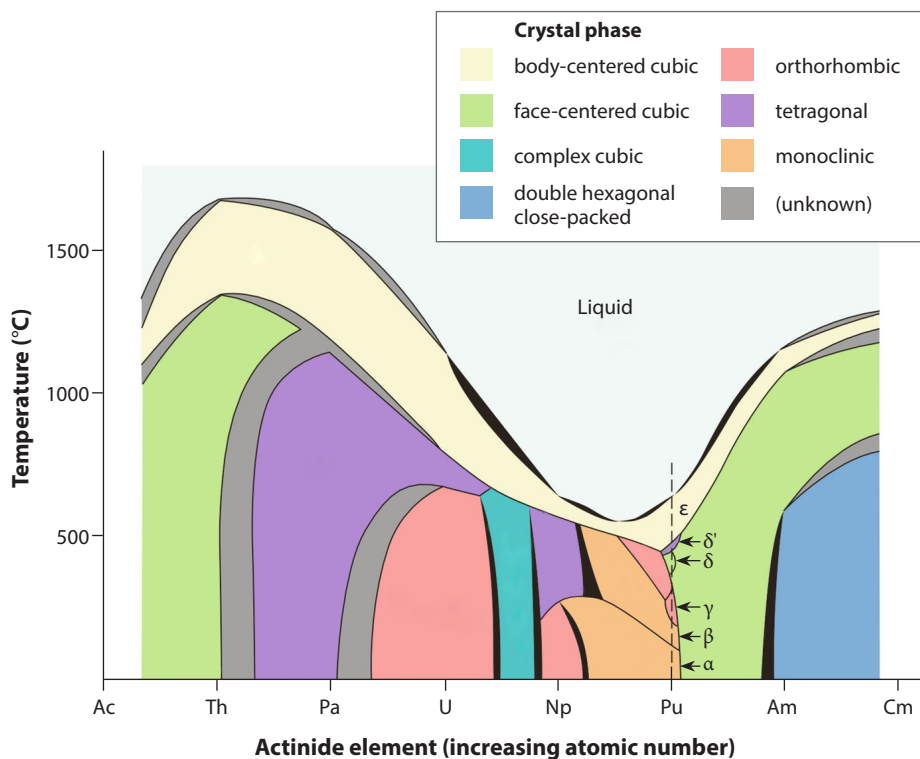
(b) The Pu temperature-pressure phase diagram shows that pressure stabilizes the metal in its least symmetric, most complex atomic arrangements, i.e., α - or β -phase monoclinic solids or a structureless liquidus.



(c) The complexity of chemical alloying for Pu-Ga is shown with varying ratios and temperatures. Six allotropes of Pu, several new binary phases, and 11 intermetallic compounds are formed.



(d) Time dependence of non- $^{239/240}\text{Pu}$ constituent concentrations in weapons-grade plutonium resultant from radioactive decay and decay daughter product ingrowth. With half-lives shorter than 100 years, the decay of ^{241}Pu and ^{238}Pu isotopes occurs rapidly compared to ^{242}Pu with a 240,000 year half-life.



Connected actinide phase diagram of binary alloys. This shows the progression of favored elemental crystal phases as the actinide group is traversed, with plutonium clearly situated at an abrupt transition—the six allotropes are shown at this transition. [Adapted from “Magnetism or Bonding: A Nearly Periodic Table of Transition Elements” by James Smith and Ed Kmetko, published in the *Journal of the Less-Common Metals* in 1983.]

Properties of plutonium

As a brief introduction to plutonium science, I present the unusual properties of plutonium in the table on the opposite page to give the reader an appreciation for the complexities of plutonium. The greatest engineering challenges arise from its notorious instability as shown in the figures below the table. Plutonium metal is unstable with respect to temperature, pressure, chemical additions, and time. The metallurgical challenges for engineering applications of plutonium are particularly great because of its instability and the myriad phase transformations it exhibits.

Plutonium defies conventional metallurgical wisdom, so we must turn to its electronic structure to gain better insight. Many of the properties described above are telltale signs of novel interactions and correlations among electrons. Boring and Smith in their 2000 article, “Plutonium condensed matter physics,” published in *Los Alamos Science*, point out that such novel interactions typically result from a competition between itinerancy (bonding electrons that form bands in metals) and localization (electrons with local moments that magnetically order at low temperature).

The actinides mark the filling of the 5f atomic subshell much like the rare earths mark the filling of the 4f subshell. Yet, the 5f electrons of the light actinides behave more like the 5d electrons of the transition metals than the 4f electrons of the rare earths. At the very beginning of the actinide series, there is little f-electron influence and, hence, one finds typical metallic crystal structures, few allotropes, and high melting points (this behavior is best illustrated in the connected phase diagram across the actinides shown above). As more f-electrons are added (up to plutonium), they participate in bonding (that is, they are itinerant, much like the d electrons in transition metals) and the crystal structures become less symmetric, the number of allotropes increases, and the melting points decrease. At americium and beyond, crystal structures typical of metals return, the number of allotropes decreases, and the

melting points rise: all indications of the f-electrons becoming localized or chemically inert, like the 4f electrons in the rare earths.

The peculiar properties of plutonium are not a single anomaly, as demonstrated in the binary alloy phase diagram, but rather the culmination of a systematic trend across the actinides. The transition between bonding and localization of the 5f electrons occurs not between plutonium and americium, but right at plutonium. In fact, atomic volume measurements show that the transition occurs between the ground-state α -phase and the elevated-temperature δ -phase.

Post-Cold War

The publication of the Los Alamos Science volumes on Challenges in Plutonium Science sparked a resurgence of interest in studying the fundamental properties of plutonium. At about the same time, we experienced a new programmatic challenge in that nuclear testing was banned by the Comprehensive Test Ban Treaty a few years after the end of the Cold War. Consequently, certifying the safety and reliability of the nuclear weapons remaining in the arsenal required a stockpile stewardship program that placed a premium on understanding plutonium better because we were no longer able to conduct the nuclear proof tests that allowed us to bridge the gap between our understanding of physics and actual weapon function.

The end of the Cold War dramatically altered the military postures of the US and Russia, allowing each to reverse the engines fueling the nuclear-weapons buildup. The nuclear arsenals of the two countries have been decreased by 85%. Both countries faced the challenge of keeping the remaining nuclear weapons stockpile safe and reliable without nuclear testing, as well as cleaning up nuclear contamination in the weapons complex and preventing the spread of nuclear weapons and nuclear terrorism.

Unexpectedly, the end of the Cold War also allowed American and Russian nuclear scientists to work together on nuclear safety and security issues, as well as fundamental science problems of common interest. One unresolved problem was the question of metastability of the fcc δ -phase in alloyed plutonium. I was able to work with Russia's premier plutonium metallurgist, Dr. Lidia Timofeeva, to clear up previous differences in Russia's favor as described in the "Tale of Two Diagrams," published in Los Alamos Science in 2000.

The next major challenge in plutonium science and technology was to understand the aging of plutonium because the end of nuclear testing and the closure of US plutonium manufacturing facilities at the Rocky Flats Plant required a lifetime extension for the plutonium components in US weapons of many decades. In addition to typical concerns of materials aging from the outside-in through surface reactions, plutonium ages from the inside-out because of the relentless deposition of energy from its alpha decay, which damages its crystal lattice and transmutes plutonium into other elements over time.

At cryogenic temperatures (4 K), lattice damage causes an apparent loss of crystallinity with long irradiation times. At room temperature, much but not all of the lattice damage is annealed out because defects produced by self-irradiation are sufficiently mobile. Small nanometer-size bubbles form quite rapidly. Much effort continues to be devoted to understanding the effect of these bubbles and other changes with age on the properties and performance of plutonium, particularly since self-irradiation may affect plutonium's delicate balance of stability with changes in temperature, pressure, or chemistry.



The author (center) on a remarkable visit to the Yongbyon nuclear complex in North Korea, August 2007.

My journey with plutonium also diversified from its scientific roots that began with a summer research internship at Los Alamos in 1965 and continued through my responsibilities for stockpile stewardship both as a scientist and laboratory director. With the dissolution of the Soviet Union, my interests also turned to assisting other countries to provide security for their inventories of plutonium, be it in military or civilian programs. These efforts took me many times to Russia, also to its former nuclear test site, now in Kazakhstan. I also had occasion to visit the Indian and Pakistani nuclear sites, and remarkable visits to North Korea's nuclear complex and its plutonium laboratories.

Plutonium, and nuclear materials in general, offer the prospects of peace and prosperity through judicious military employment and civilian use, such as nuclear electricity, nuclear medicine, and nuclear batteries. However, they also hold the potential seeds of war and disaster if not managed properly. We depend on the next generation to be able to manage this balance so that we can look back at the 100th anniversary of the Manhattan Project and be able to declare it a success.

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5. S.S. Hecker, L.F. Timofeeva, "A Tale of Two Diagrams," *Los Alamos Science*, 2000, 26, Los Alamos National Laboratory, 244.



David L. Clark

Dr. Clark is a Scientist at Los Alamos National Laboratory and Director of the Laboratory's National Security Education Center. His work has included Solubility Task for the Yucca Mountain Project (1993–1997), Source Term Test Program for the WIPP license application (1996–1997), program manager for plutonium aging and pit lifetime assessments (1998–2003), technical advisor for environmental stewardship including the Rocky Flats cleanup and closure (1995–2005), closure of High-Level Waste tanks at the Savannah River Site (2011), and technical advisor to the DOE High Level Waste Corporate Board (2009–2011). Dr. Clark served as inaugural Director of the Glenn T. Seaborg Institute for Transactinium Science (1997–2009).

Chemistry Challenges for the Manhattan Project and Beyond

David L. Clark

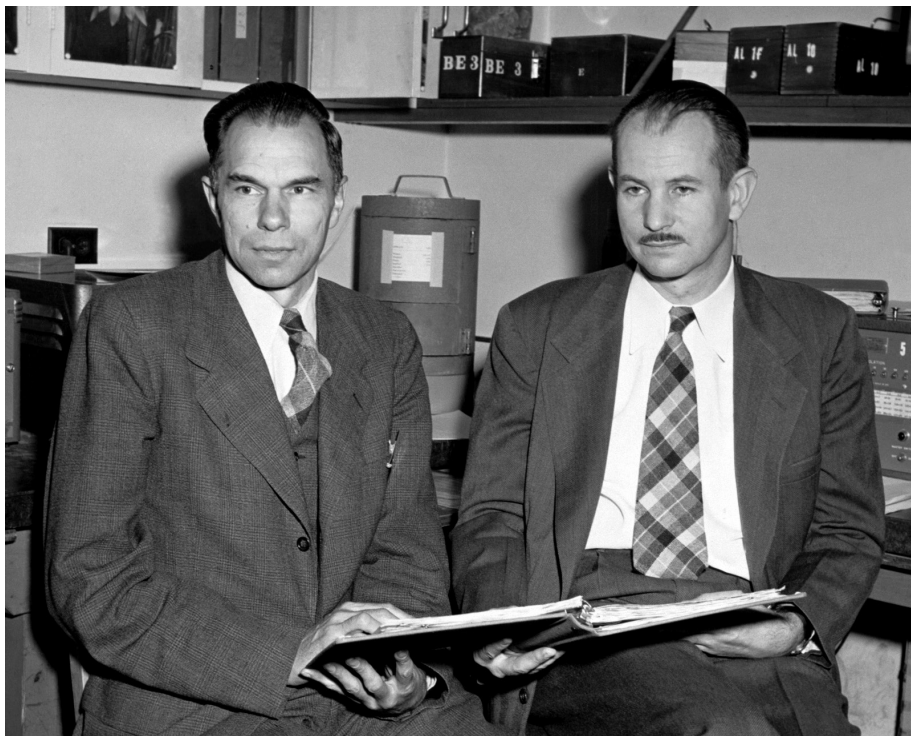
National Security Education Center, Los Alamos National Laboratory, Los Alamos, New Mexico

The important isotope ^{239}Pu was discovered in 1941 as the decay product of ^{239}Np produced with neutrons from a cyclotron. The importance of plutonium comes from its fission properties and the capability of being produced in large quantities. In 1941, Segré, Kennedy, Wahl, and Seaborg bombarded a 1.2 kg sample of uranyl nitrate with 16 MeV neutrons for two days. The uranyl species was extracted into hydrocarbon solvent and the ^{239}Np product was separated into an aqueous phase using an oxidation-reduction and precipitation process with La and Ce fluoride carriers. Measurement of the radioactive decay demonstrated that they had produced 0.5 μg of ^{239}Pu . On March 28, 1941, they used that 0.5 μg sample to demonstrate that ^{239}Pu undergoes slow neutron-induced fission with a fission cross-section for ^{239}Pu that was approximately 50% greater than for ^{235}U , agreeing remarkably well with more accurate values determined later. This observation that ^{239}Pu was fissionable with slow neutrons provided the information that formed the basis for the US wartime Plutonium Project of the Manhattan Engineer District (MED) centered at the Metallurgical (“Met”) Laboratory of the University of Chicago. Most of these early studies were carried out under a self-imposed cover of secrecy due to the potential military applications of plutonium and were not published until after World War II.

Chemistry challenges

Only tracer quantities of plutonium existed at the beginning of the Manhattan Project, therefore the initial chemistry challenges were to: develop a large-scale production method for plutonium; develop a method for its chemical separation and purification; scale up the separations from micro- to kilograms. Fermi solved the first problem by demonstrating that uranium would undergo a nuclear chain reaction on December 2, 1942: the neutrons produced in the reaction create plutonium. The solution to the second and third problems required determining the chemical properties of plutonium so that a large-scale separations plant could be designed to separate the enormous quantity of fission products and uranium. Berkeley professor Glenn Seaborg led a large group of chemists and engineers to solve this problem.

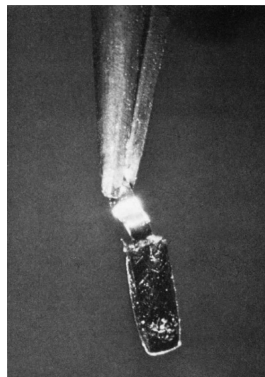
The key to plutonium separation was the oxidation-reduction cycle, in which plutonium is “carried” in its lower oxidation state(s) by chemical precipitates and not carried when it is present in higher oxidation states. Plutonium therefore becomes separated from the fission products, which do not exhibit these differences in carrying behavior. These carrier techniques had been developed for use with trace quantities of newly discovered atoms. It was unclear at the time if these techniques could be scaled up and actually used in a chemical separations plant. An entirely new effort in ultramicrochemistry was developed and led by Burris Cunningham to determine the chemical properties of plutonium because they only had sub-microgram quantities at the time. Hundreds of pounds of uranium were bombarded with neutrons at the Washington University cyclotron, and chemically separated down to 2.77 μg as the first weighable sample of plutonium on September 10, 1942.



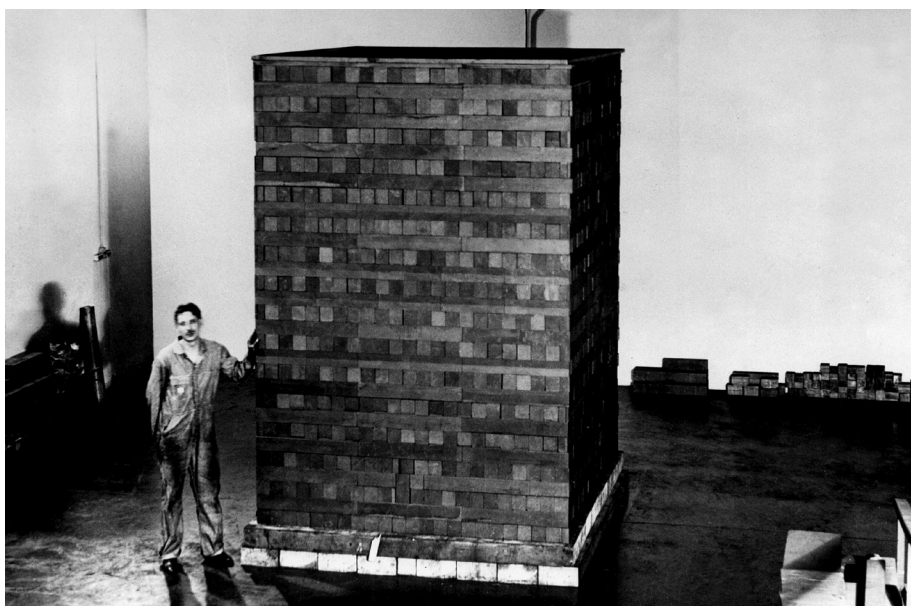
Glenn Seaborg (left) and Edwin McMillan (right) were jointly awarded the 1951 Nobel Prize in Chemistry for discovering the transuranium elements including plutonium.

“We kept it secret voluntarily and when we reported this to the people in Washington, this really became the basis for the plutonium part of the Manhattan Project, the atomic bomb project”

– Glenn Seaborg on the discovery of plutonium

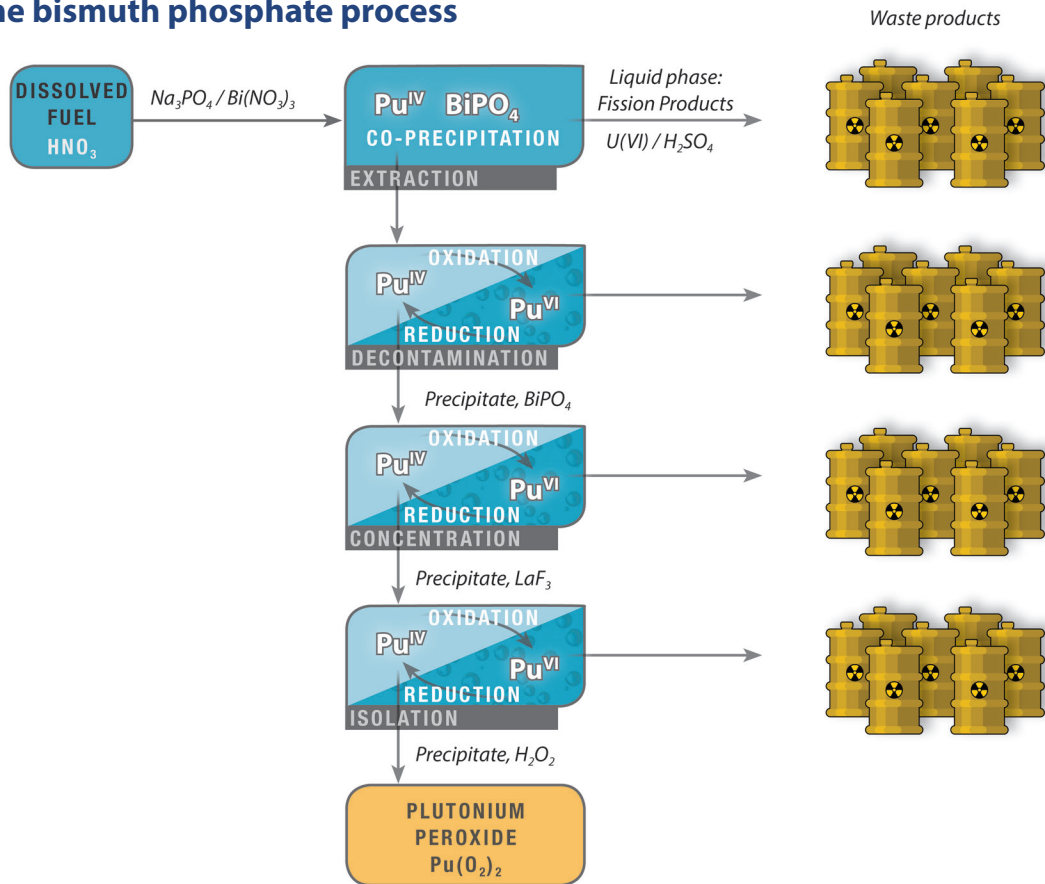


The first isolated sample of plutonium large enough to be weighed, September 10, 1942 (2.77 μg ; 20 \times magnification on a platinum pan). The sample is of the oxide, PuO_2 , which is visible as a thin crust on the pan towards the bottom of the photograph.



A worker stands with one of at least 29 experimental piles that were constructed in 1942 under the West Stands of Stagg Field, Chicago, IL. Controlled nuclear fission of uranium was achieved using these piles by a team led by Enrico Fermi on December 2, 1942. Without this technological advancement, the large-scale production of plutonium from uranium, necessary for the Manhattan Project, would have been impossible.

The bismuth phosphate process



Above: The bismuth phosphate process. This was used for the first large-scale purification of plutonium from neutron-irradiated uranium at the Hanford site during the Manhattan Project and up until the 1950s. The key to this process is the use of bismuth phosphate as a "carrier" to precipitate Pu(IV) salts in nitric acid. Oxidation to Pu(VI) allows further separation of the waste, and this oxidation-reduction-precipitation cycle is repeated many times over. It produces a large volume of waste, and consumes large quantities of chemicals.

The bismuth phosphate process

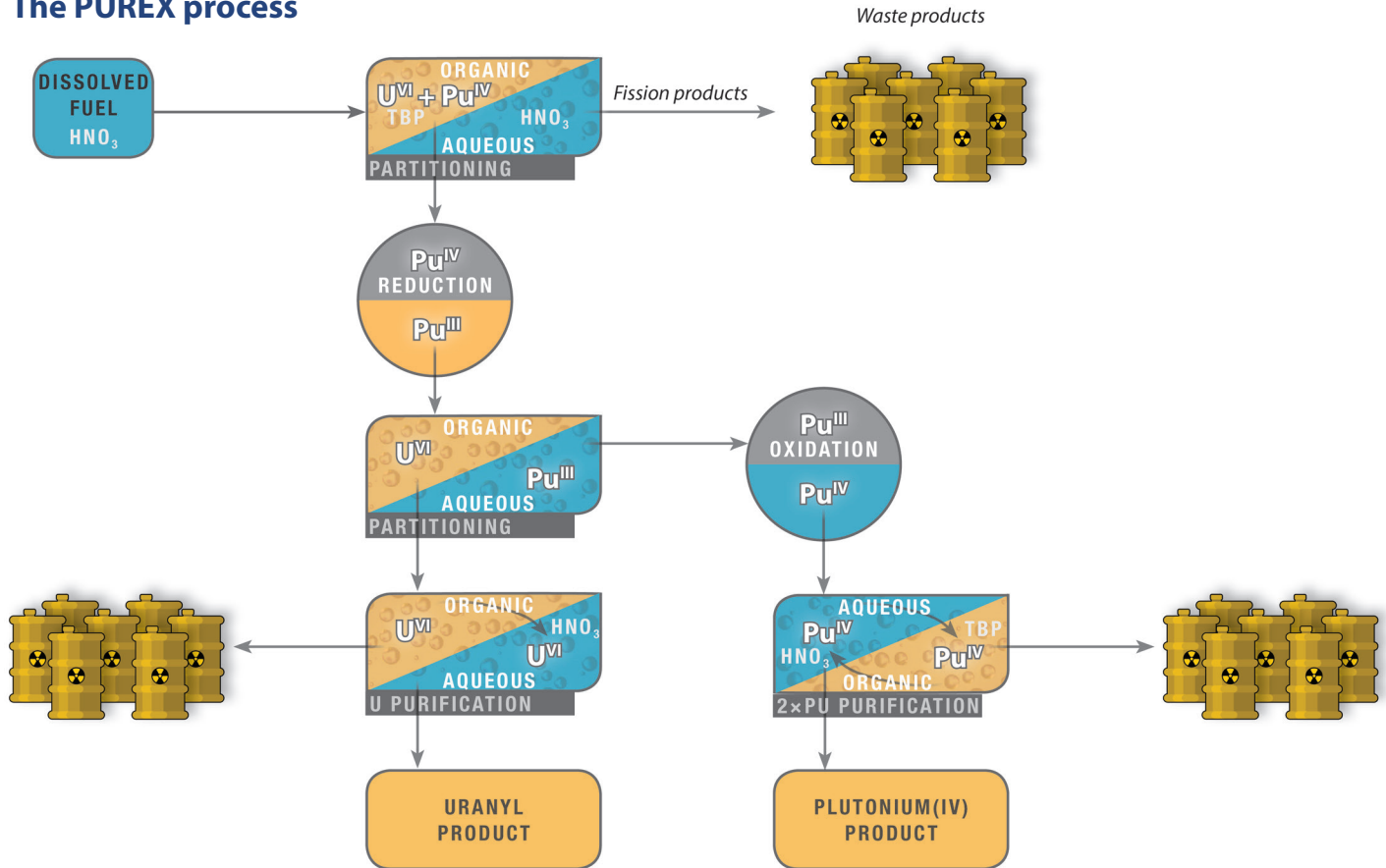
The Seaborg team had to find a way of separating plutonium in high yield and purity from the many tons of uranium in which plutonium was present at a maximum concentration of only 250–300 ppm. Because of these low concentrations, compounds of plutonium could not be precipitated directly, and any precipitation-separation process had to be based on coprecipitation with "carriers" for plutonium. Bismuth(III) phosphate was chosen as the carrier. In addition, the highly radioactive fission products had to be separated to less than one part in 10⁷ of the original plutonium. This rigid requirement was necessary so that separated plutonium was safe to handle. Without separation from the fission products, the plutonium from each ton of uranium would have more than 10⁵ Ci of energetic gamma radiation.

The key to the bismuth phosphate process is that it quantitatively carries Pu(IV) from acid solution but does not carry Pu(VI). Unfortunately, the process suffers from the batch nature of operations, the large amounts of chemicals used, and large amounts of waste. The Hanford site began construction of tank farms in the 1940s and 1950s to hold these large quantities of waste.

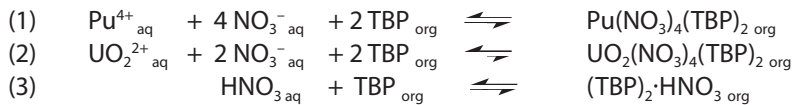
After the Manhattan Project: PUREX, the game changer

During the Cold War, the PUREX (Plutonium Uranium Redox EXtraction) solvent extraction process revolutionized plutonium separations. In solvent extraction, the species to be separated is transferred between two immiscible or partially-miscible phases, such as water and a nonpolar organic phase. The process works by selectively complexing the actinide species of interest, decreasing its solubility in water while simultaneously increasing its solubility in the organic phase. By far the most important

The PUREX process



and widely used neutral extractant is tributylphosphate (TBP). It complexes with the actinide elements Th, U, Np, and Pu by forming inner sphere chemical bonds to the actinide metal atom via the phosphoryl P=O bond. The important reactions for UO_2^{2+} and Pu^{4+} are shown below:



The reactions are equilibrium reactions, therefore the ratio of products, and thus the degree of extraction, can be increased by increasing the concentration of TBP or NO_3^- in the organic and aqueous phases, respectively. These extraction equilibria are the basis of the PUREX process, used almost exclusively worldwide in all modern reprocessing of spent nuclear fuel. In the PUREX process, irradiated UO_2 fuel is dissolved in HNO_3 , with uranium being oxidized to $\text{UO}_2(\text{NO}_3)_2$ and plutonium to $\text{Pu}(\text{NO}_3)_4$. A solution of TBP in a high-boiling-point organic solvent such as n-dodecane is used to selectively extract hexavalent $\text{UO}_2(\text{NO}_3)_2$ and tetravalent $\text{Pu}(\text{NO}_3)_4$ from the other actinide and fission product nitrates in the aqueous phase. In the second extraction container, a TBP solution is contacted with a dilute HNO_3 solution containing a reducing agent such as ferrous sulfamate, which reduces plutonium to Pu(III), but leaves the uranium as U(VI). Plutonium then transfers back to the aqueous phase leaving uranium in the organic phase. The uranium is stripped from the organic phase using water.

Above: The PUREX process, used since the 1950s as the primary method of purifying plutonium. An organic solution of tributylphosphate (TBP) is used to extract U(VI) and Pu(IV) from aqueous waste fission products; plutonium is subsequently separated using a reducing agent, which brings it back into the aqueous phase.



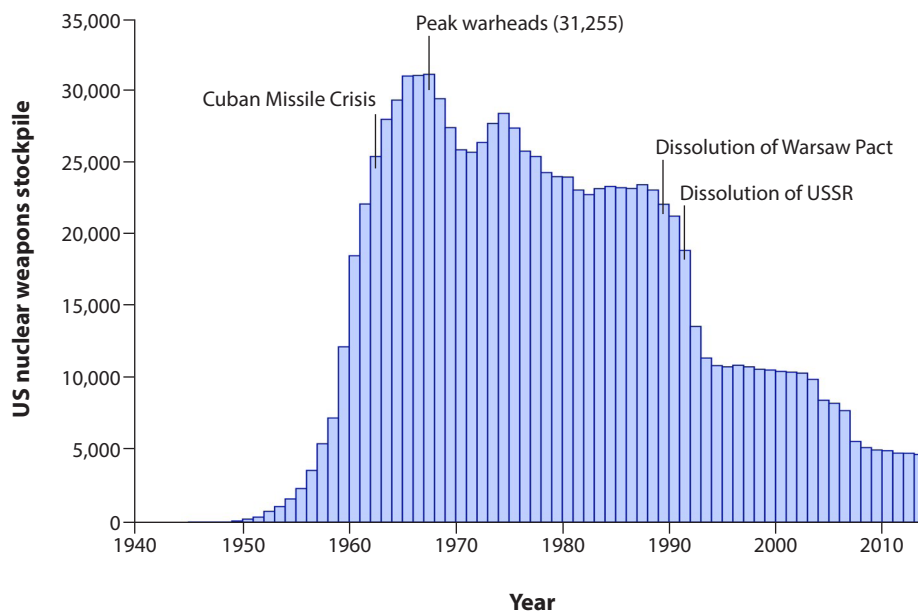
At Hanford and the Savannah River Site (SRS) massive underground storage tanks were built to accommodate high-level waste (Hanford sites shown above). These 226 tanks contain 575 million curies in 91 million gallons of sludge, liquid, and solid waste. Today, the tank farm at the SRS alone costs the US taxpayer \$1 million every day to operate. As part of the DOE cleanup effort, the legacy waste is being treated via a vitrification process, i.e., turning the sludge into a more stable glass form at high temperatures. This is then packaged into steel containers for decontamination and disposal. Estimates for life-cycle costs reach nearly \$250 billion with completion of the cleanup of SRS and Hanford tank farms by 2060–2070.



The Hanford PUREX plant was authorized in 1953, and hot operations began in January of 1956. The initial processing rate was 200 MT/U/month. PUREX capacity soared and by 1961, PUREX was processing 800 MT/U/month. Although the PUREX waste-to-product ratio was much lower than other processing plants, the need for waste disposal soared. Hanford responded with many different campaigns to build new waste tank farms to store the highly radioactive waste.

The tank waste legacy

Managing and treating the tank wastes stored in the farms of aging underground tanks at the Savannah River Site (SRS) and Hanford has been a grand challenge for the DOE Office of Environmental Management (EM) mission, posing a significant threat to environment, safety, and health. The tank farms at SRS and



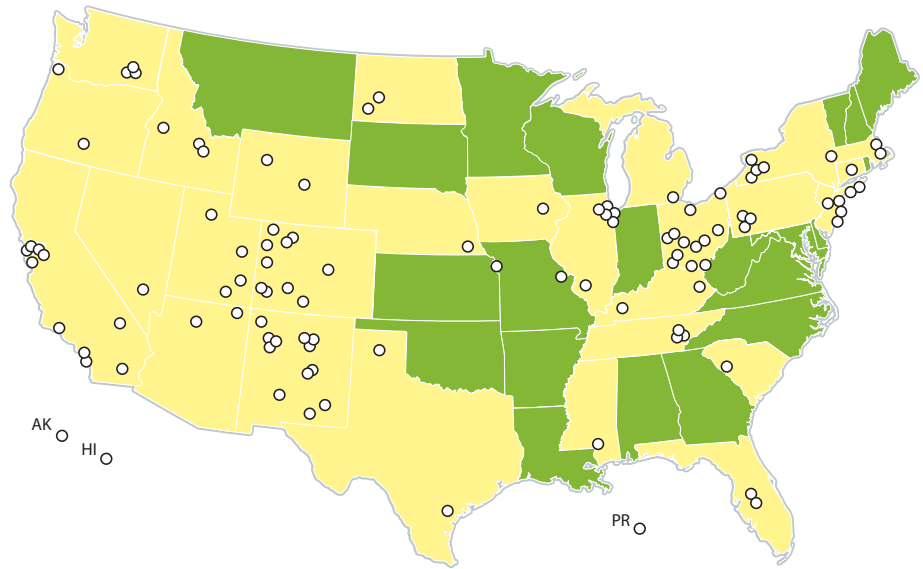
An enormous Cold War buildup of the U.S. nuclear stockpile drove plutonium production. [Source: Dept. of State; Transparency in the U.S. Nuclear Weapons Stockpile, 2015]

Hanford contained the majority of the Department of Energy (DOE) tank waste inventory with approximately 575 million curies of radioactive materials in 91 million gallons of sludge, liquid, and solid waste stored in 226 underground tanks. The majority of activity is stored in SRS tanks (400 million Ci), while the largest volume (53 million gallons) are stored in Hanford tanks (see figure on page opposite). The costs for managing the tank farms are enormous with about \$1 million per day for tanks at SRS and life-cycle costs in the billions of dollars. Estimates for life-cycle costs reach nearly \$250 billion with completion of the cleanup of SRS and Hanford tank farms at the latest by 2062. Although EM has made significant progress in its cleanup mission, the majority of the tank wastes remain untreated. Only seven tanks have been emptied and two closed at SRS; no tanks have been closed at Hanford. Given the enormous task to retrieve, treat, and dispose of the large volumes of highly complex and highly radioactive tank wastes, opportunities exist to invest in the development of advanced technologies and scientific understanding of tank waste issues that can accelerate the cleanup mission and reduce life-cycle costs.

Savannah River Site

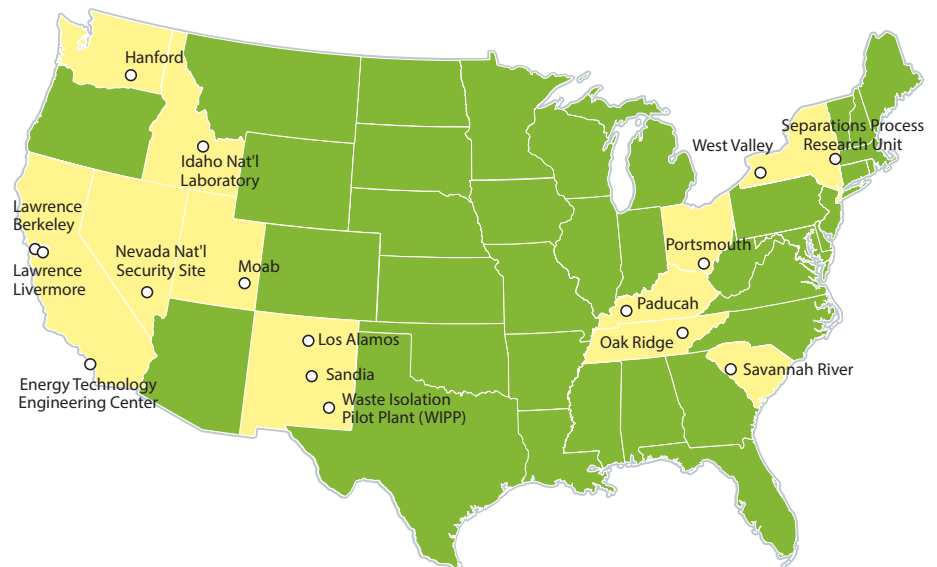
The Savannah River Plant was built and operated as a second production site for plutonium and other nuclear materials producing well over 100 million gallons of radioactive waste stored in underground tanks. The main process used for treating spent nuclear fuel and separating plutonium was PUREX, described on the previous page. The wastes were made alkaline for storage in carbon steel tanks, producing an insoluble sludge consisting of actinide and fission products and a supernatant liquid containing the majority of the ¹³⁷Cs. To date, SRS underground tanks received about 140 million gallons of radioactive waste, which was reduced to approximately 36 million gallons by evaporation. The radioactive waste is currently stored in 49 underground tanks containing approximately 350 million curies of radioactive material. The SRS tanks reportedly contain about 16.9 million gallons of supernate, three million gallons of sludge, and 16.6 million gallons of salt cake. Of the underground tanks, 27 have full secondary containment in compliance with the site's Federal Facility Agreement (FFA). The remaining 22 tanks have only one or partial second containment and, therefore, are considered non-compliant tanks.

1989



In 1989, the Department of Energy (DOE) weapons complex had 107 contaminated sites in 35 states, spanning 3,100 square miles (*top*). By 2019, it has reduced its footprint by 90% to less than 300 square miles to 16 sites in 11 states (*bottom*).

2019



Some of the SRS waste has been treated by incorporating the radioactive components into borosilicate glass at the Defense Waste Processing Facility (DWPF) and decontaminated supernate into a cement-based waste form referred to as saltstone. In 2008, the DOE entered into a contract with Savannah River Remediation LLC to accelerate closure of the tanks, and requires that all waste must be removed from all tanks by 2028. As of 2016, the DWPF had produced 4,000 glass canisters. Final closure and grouting of the final H-area East Hill tank is scheduled for fiscal year 2032.

Hanford Site

The Hanford Reservation was the first industrial-scale plutonium production site in the world including multiple reactors and reprocessing facilities. Plutonium and spent fuel were processed in five reprocessing plants, creating large volumes of liquid and solid radioactive wastes. Past waste disposal management involved disposal into the environment and storage in large underground tanks. The Hanford tanks contain 53 million gallons of highly radioactive and chemical waste, only about 10% of the originally generated waste volume. The high-level waste (HLW) is stored in 177 single- and double-shell tanks containing approximately 175 million curies of radioactive constituents. Nearly 70 single-shell tanks have or are suspected to have leaked up to 1.5 million gallons of waste into the surrounding soil, while none of the 28 newer, double-shell tanks have lost their integrity.

Most of the waste removal and tank closures have yet to be performed, awaiting the operation of the large Hanford Tank Waste Treatment and Immobilization Plant (WTP). The plant will use vitrification technology, which involves blending the waste with glass-forming materials and heating it to 1,150°C. The molten glass mixture is then poured into stainless steel canisters to cool and solidify. In this glass form, the waste is stable and impervious to the environment, and its radioactivity will safely dissipate over hundreds to thousands of years. The plant is scheduled to begin operations in 2023, but has been plagued by setbacks.

Summary

The creation of atomic weapons and the buildup of the US Cold War nuclear arsenal has left an environmental cleanup legacy of enormous cost and scope—the largest environmental cleanup program in the world. Through science, technology, and engineering, the US has developed innovative solutions and reduced the legacy footprint by 90% to less than 300 square miles at 16 sites in 11 states—no other country has done this. Legacy cleanup is necessary to transform the US nuclear weapons complex and provide stewardship of a smaller US stockpile. Future challenges at Hanford and SRS will give the US experience in HLW treatment, essential for managing the legacy of future wastes and spent nuclear fuel (a separate challenge). Finally, integrating worker safety and environmental protection into processes and facilities is an essential element of maintaining a modern stockpile.

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4. GAO-19-223, Report to Congress, "Nuclear Waste Cleanup," Feb 2019.
5. GAO-19-460T, Report to Congress, "Environmental Liability Continues to Grow, and Significant Management Challenges Remain for Cleanup Efforts," May 2019.
6. GAO-18-241, Report to Congress, "Hanford Waste Treatment Plant," April, 2018.



James L. Smith

Dr. Smith started working for Los Alamos National Laboratory in 1973 as part of the Physical Metallurgy Group. Over his forty-year career at Los Alamos, Smith worked on low-temperature physics, superconductivity, magnetism, and actinide materials. Specifically, Smith studied the relationship between superconductivity and magnetism and helped pioneer the field of heavy fermion superconductivity. Over the course of his career, Smith authored over 400 papers. In 1982, Smith became a Laboratory Fellow at Los Alamos. He retired in 2013, and currently serves on the Atomic Heritage Foundation's Advisory Committee.

Physics Underpinnings: A Perspective

James L. Smith

Los Alamos National Laboratory, Los Alamos, New Mexico

The following is a transcription of Dr. Smith's oral presentation.

The neutron was discovered in 1932 by James Chadwick. The name “neutron” was already taken (derived from its neutral charge), and so Enrico Fermi changed the name of the other particle to “neutrino” by using an Italian diminutive ending on neutron. Fermi began using neutrons from radium- or radon-beryllium sources to transmute the nuclei of atoms and was awarded the Nobel Prize in 1938 for creating new radioactive elements.

Radioactive elements are now in great demand as medical isotopes and are produced at the Los Alamos Neutron Science Center (LANSCÉ) by diverting part of the proton beam to transmute the nuclei of atoms before it reaches its full energy. Fermi thought he had found the element after uranium, but it sure had a lot of funny radiation: fission fragments, we know now. In 1938, Hahn and Strassman reported that they had split the uranium nucleus, and immediately after that Meitner and Frisch explained that neutrons and a great deal of energy were released, coining the term “fission”. The cognoscenti immediately understood that a bomb was possible, and the only question was how difficult that might be.

The US got off to a slow start, but Los Alamos became part of the Manhattan Project in 1943. That same year, Sig Hecker and I were born 29 days apart. My first language was English, and his was German. Radios took minutes to warm up; calculations were done on slide rules; food was organic because insecticides and herbicides had not been invented. There were important materials discoveries during the Manhattan Project. The role of the chemists, metallurgists, and solid-state physicists was to provide the framework for the nuclear physics to play out. Materials had to be purified and formed into shapes. On the opposite page is shown Larry Litz's notebook page from D-Day (June 6, 1944). By the time people in the US got to work that day, it was already on the radio that the invasion of France was underway. You can see that Larry was worrying about outgassing and light-metal impurities in his samples. Vern Struebing meanwhile cast the plutonium for Trinity and Combat. Everyone used induction heating for melting materials, and later when Vern showed me how to use it, he had an optical pyrometer with a shoulder strap on it. I had never seen lab equipment with such a strap; it was from the Manhattan Project and was of a design used in steel mills of the day.

The Manhattan Project is historically recognizable as the first metallurgical study and application of actinides. If you arrange the elements of the f- and d-electron series in order of increasing orbital size and overlap (see figure opposite), a pattern emerges. While s- and p-electron solids are simpler to understand, the filling of f and d electronic shells leads to gradual changes in materials properties as the atomic volumes contract along the series. If the electrons overlap in bonding the elements may become superconducting (shown in blue), whereas isolated localized electrons in a partially-filled shell can possess a magnetic moment and order magnetically at low enough temperatures (shown in yellow). Thus, these regions are simple enough

for B with no change, 1325°C. 10' (steady
 time), slight darkening on tube at ~1200-1300°C.
 On removal, material appeared in essentially
 the same shape, black metallic cob, adding
 to the crucible very strongly.

6.6.44 D-day
 Experiment # 7.

1. Oxidant, sulfide crucible degressed at 1600°C.
 " " " " " " 1500°C.

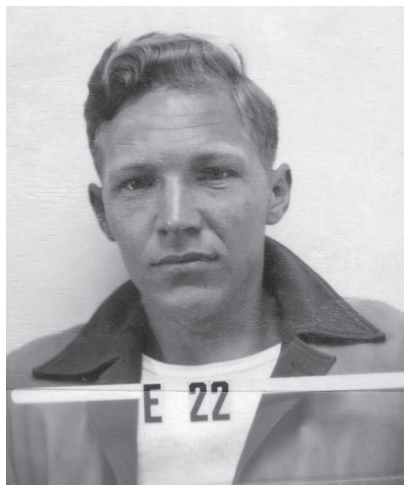
2. Sample # 1742 - Baker. Had been in trans-
 formation m.p. experiment. 675 mg. were
 placed in the Be₂S₃ crucible. Heated at 900-950°C.
 for 10 min. to 10:30. Cr. about 1 min. Remelted
 metal formed nice crystal, wt. = 675 mg.
 Plutonium 1.79%. Part of thermocouple ~~was~~
 included in the button for m.p. exp.

Anal of 1742: U = 25 ppm, Ni = 20 ppm, Be = 1 ppm
 Na = 50 ppm (20), Mg = 2 ppm, Al = 10 ppm, K = 20 ppm,
 Ca = 200 ppm

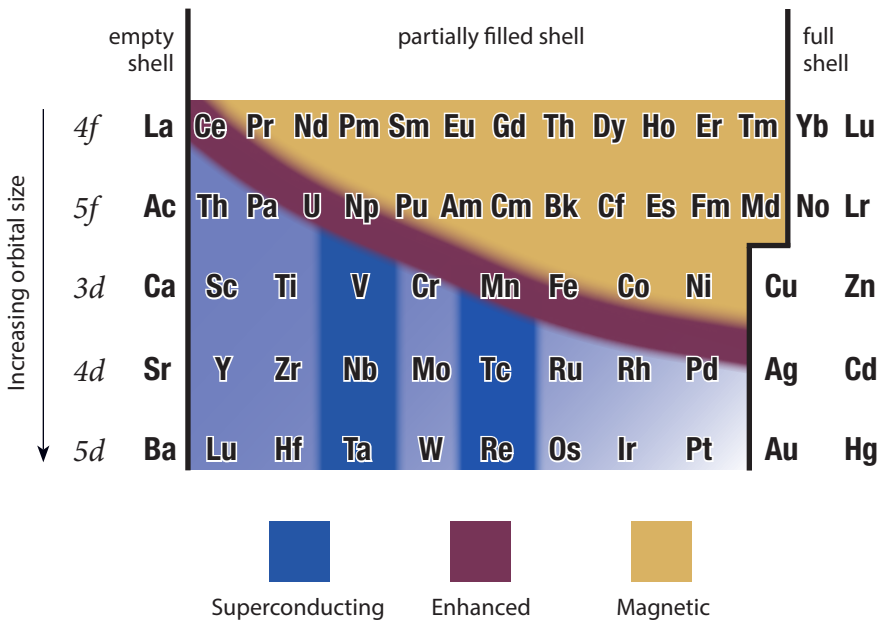
Anal of # 7791-1
 Ni = 1 ppm
 Mg = 10 "
 Al = 2 "
 K = 40 "
 Ca = 25 "



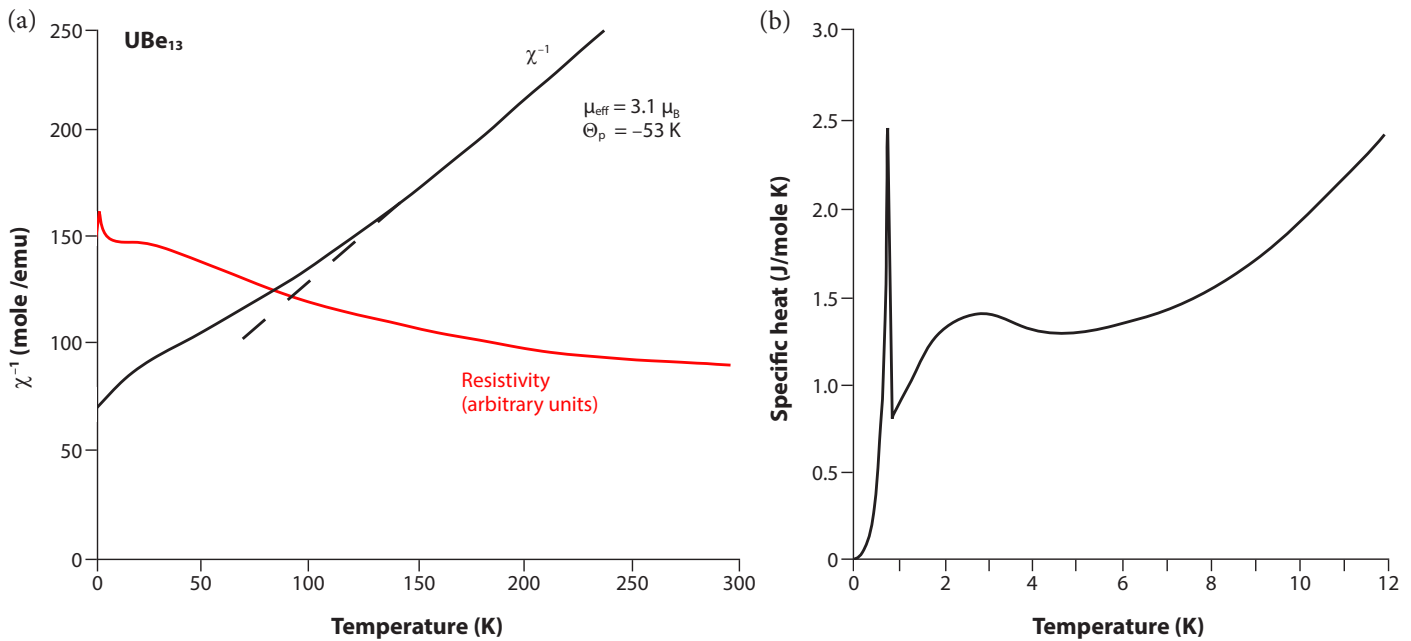
Lawrence Litz began working on radioactivity at the Metallurgical Laboratory at the University of Chicago. He was then transferred to Los Alamos during the Manhattan Project, where he developed the casting process for the plutonium hemispheres in the atomic bombs. He was the first person to see metallic plutonium. Left: Extract from his laboratory notebook on D-Day (June 6, 1944).



Vernon Struebing was a chemist who worked at Los Alamos as a civil scientist during the Manhattan Project. His work was focused on plutonium metallurgy; he cast the plutonium for Trinity and Combat. After the war, Struebing worked in the Plutonium Physical Metallurgy Group, which studied the physical properties of plutonium.



An arrangement of the elements with the position of the d- and f-blocks reversed, in order of increasing radial size of valence orbitals. It shows the crossover between electron bonding behavior and magnetic moment formation. [Adapted from "Magnetism or Bonding: A Nearly Periodic Table of Transition Elements" by James Smith and Ed Kmetko, published in the Journal of the Less-Common Metals in 1983.]



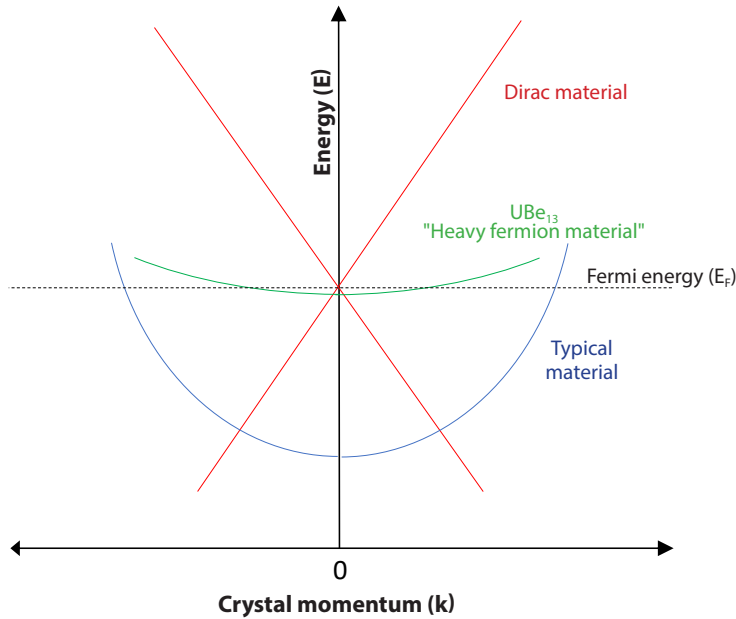
Above: The actinide binary compound UBe_{13} is an unconventional low-temperature superconductor, and one of the early materials of its type to be studied. It is still not fully understood. Some of its unique properties are shown in these two graphs. (a) The plot of inverse magnetic susceptibility versus temperature should show a linear relationship for a material which obeys the Curie-Weiss law. A deviation from this exists below liquid nitrogen temperatures, along with increasing resistivity, which counter-intuitively peaks when the material becomes superconducting.

(b) The plot of heat capacity shows a peak at the superconducting temperature. [Adapted from “ UBe_{13} : An Unconventional Actinide Superconductor” by Ott, Rudigier, Fisk, and Smith, published in Physical Review Letters in 1983.]

to understand, but in the area where the electrons are intermediate between the two states, interesting behavior occurs (shown in purple, “enhanced”). The material properties become quite sensitive to perturbations such as temperature, pressure, impurities, magnetic fields, and stress. Therefore, these metals have multiple crystal structures, variable properties, and are pyrophoric, i.e., make sparks when struck. The figure in Sig Hecker’s article on p17 shows the actinide row that displays these properties well. These are the binary alloy phase diagrams of pairs plotted in a series. This series figure shows the way the melting points plummet; the crystal structures multiply and then go back to normal as the f electrons cross over from bonding to localized behavior. It also shows that plutonium, the element of most interest in the Manhattan Project, resides at this intersection.

Strange things can emerge in materials. Consider the binary compound UBe_{13} . The uranium atoms are spread out by the non-interacting beryllium atoms and so should possess magnetic moments. As seen in the magnetic susceptibility plot above, it has a moment above liquid-nitrogen temperature (77 K). But everything else is crazy. The heat capacity looks magnetic, but goes superconducting at about 1 K. The superconducting electrons behave as though their masses were a thousand times larger than that of a bare electron. These electrons scarcely want to move even though they are superconducting. This so-called “heavy-fermion” superconductor and other similar actinide compounds are not completely understood.

In 1987, superconductivity was found above liquid-nitrogen temperature. Edward Teller, a Manhattan Project alumni, co-inventor of the hydrogen bomb, and co-founder of Lawrence Livermore National Laboratory, wanted someone to teach him superconductivity. He looked around the two labs and picked me as his teacher. We spent about 100 hours together and became friends. He was a good student. Currently, the superconducting transition temperatures are approaching room temperature (at present the record high is 250 K). I have no doubt they will get to room temperature, but the difficulty will be making these superconductors useful (for example, when superconductivity is only observed under elevated pressures). This type of high-temperature superconductivity is not fully understood.



Schematic representation of energy dispersion curves for electrons in a Dirac material, a typical material (i.e., copper), and UBe_{13} . These dispersion curves represent the relationship between an electron's energy (E) and its crystal momentum ($k = p/\hbar$, where p is its momentum). Electronic behavior in a Dirac material and UBe_{13} deviates from that in a typical material due to crystal structure, spin, electronic correlations, and other physics.



Edward Teller and the author, May 23, 1989, Huntsville, AL. Teller was an early member of the Manhattan Project, and proposed the solid pit implosion design which was successful. He co-founded Lawrence Livermore National Laboratory with Ernest Lawrence, and was both its director and associate director for many years.

Superconductors are examples of emergent phenomena, namely complex and unexpected behavior arising from simple things. The Gulf Stream is an example of emergent phenomena from oceanography. So what does the future hold for emergent phenomena? Paul Dirac performed mathematical calculations defining a Dirac material in which electron energy is linear in the absolute value on momentum, unlike a typical materials electron energy which goes as $p^2/2m$ ($p =$ momentum, $m =$ mass). This seems quite curious to have electrons behaving like light. However, Dirac materials have been realized recently in solids. Here the spin would still give a degeneracy, but it is still not that simple. In Weyl materials, only one spin is associated with a particle. These materials may lead to more powerful computers, which in turn may help us figure out what is going on with over half of the matter in the universe that we cannot see or understand. (It is our meeting organizer Alexander "Sasha" Balatsky who wrote this, and so ask him, not me, about it!)

Why do we still care about nuclear weapons? We cannot just leave them somewhere because they are radioactive and contain explosives. I do subscribe to the view that their existence has prevented another world war. If we get a bit smarter our costs can be reduced substantially. And as long as we do not resume testing them, they will slowly get less important. A more active plan aimed at eliminating nuclear weapons is far more desirable, but the non-testing default is a minimum possibility.



Galya Balatsky

Dr. Balatsky is a member of the Intelligence and Systems Analysis group (A-2) and has been with Los Alamos National Laboratory for nearly 20 years. She has participated in several DOE nuclear nonproliferation projects and has worked with the Institute of Nuclear Materials Management since 2004, promoting research and development, and led workshops such as Reducing Risk from Radioactive and Nuclear Materials.



Parrish Staples

Dr. Staples has over 25 years of experience with nuclear industry and is currently working as a senior consultant for a variety of programs throughout the DOE complex. He completed his federal career as the Director of the Domestic Uranium Enrichment Program and was Director of the Office of European and African Threat Reduction including as the Director of the Reactor Conversion and Mo-99 production programs.

From Manhattan to Nonproliferation: What Will Be the US Role in Future Nonproliferation?

Galya Balatsky, Parrish Staples

Center for Intelligence & Systems Analysis, Los Alamos National Laboratory, Los Alamos, New Mexico; Staples Science & Policy Consulting LLC, Las Vegas, Nevada

To date, the established rules, regulations, and international consensus regarding nonproliferation have successfully overcome a variety of issues related to the spread of nuclear weapons and nuclear terrorism. Looking into the future, will the US remain a leader in nonproliferation and other security-related areas when the interest in US civilian nuclear projects has been diminishing? With growing global populations comes the growing need for energy. And with concerns over climate change, many countries consider nuclear energy as a preferred way to attain their energy needs. The countries who want nuclear technologies and nuclear-produced energy have a right to develop them but they need to do it in a peaceful, safe, and secure manner. The challenge is how to introduce and implement sophisticated nuclear technologies in “newcomer” countries, i.e., those lacking well-established industrial bases. How best to ensure nuclear technologies are proliferation-resistant? Our opinion is that it is important to be proactive and flexible.

The power of nuclear weapons became known after Hiroshima and Nagasaki, and the US leadership was concerned this information may end up in the wrong hands. President Truman signed the Atomic Energy Act in 1946 that “conserves and restricts the use of atomic energy for the national defense.” In spite of this policy of secrecy, knowledge was spreading: the USSR tested its first atomic bomb in 1949 and then in 1952 Great Britain performed their test. Under these circumstances, the decision was made to adopt a policy of controlling nuclear information through cooperation; the program “Atoms for Peace” was born and the International Atomic Energy Agency (IAEA) was established in 1957. The IAEA was mandated “to accelerate and enlarge the contribution of atomic energy to peace, health, and prosperity throughout the world” and ensure that it is not used “to further any military purpose.”

Nonproliferation efforts were later enhanced by the adoption of the Non-Proliferation of Nuclear Weapons Treaty (NPT) which came into force in 1970. The Treaty affirmed the benefits of peaceful applications of nuclear technology. The NPT obliged nuclear weapon states not to transfer nuclear weapons nor other nuclear explosive devices to any recipient, as well as not to assist nor encourage non-nuclear weapon states to manufacture or acquire nuclear weapons or nuclear explosive devices. It also places obligations on non-nuclear weapon states not to receive nuclear weapons or other nuclear explosive devices, and in addition not to manufacture nor acquire nuclear weapons or other nuclear explosive devices and not to seek or receive assistance for such. In addition, the NPT requires non-nuclear weapon states to accept safeguards, administered by the IAEA, and defines nuclear weapons states.

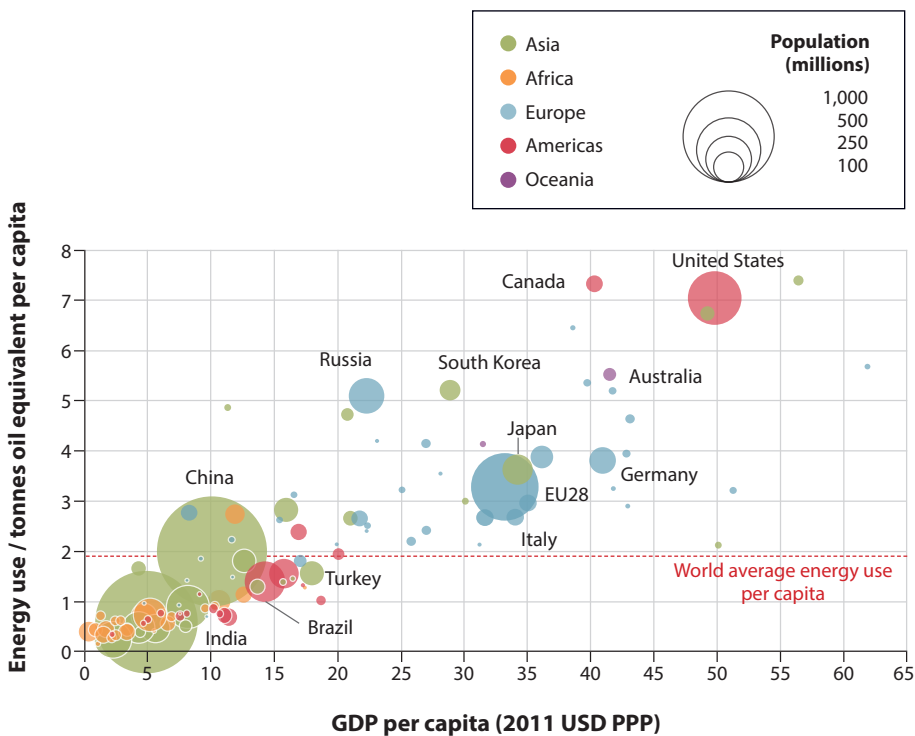
IAEA Safeguards

Civil nuclear energy activities rely on facilities and technologies that can also be used in nuclear weapons programs. The main goal of Safeguards is to monitor and verify that states do not divert materials to nuclear weapons programs. Currently, the IAEA has comprehensive safeguards agreements with 175 countries and more than 3,000 verifications were performed in 2018.

Over the years, Safeguards has been strengthened and other international instruments were added to enhance nuclear safety and security globally. The Nuclear Suppliers Group was established in 1974. The events of 9/11 brought the security of radioactive sources into focus and required countries to address the nuclear terrorist threat. The administration of President Obama held Nuclear Security Summits to improve the security of nuclear materials and generate stronger international support for nuclear security.

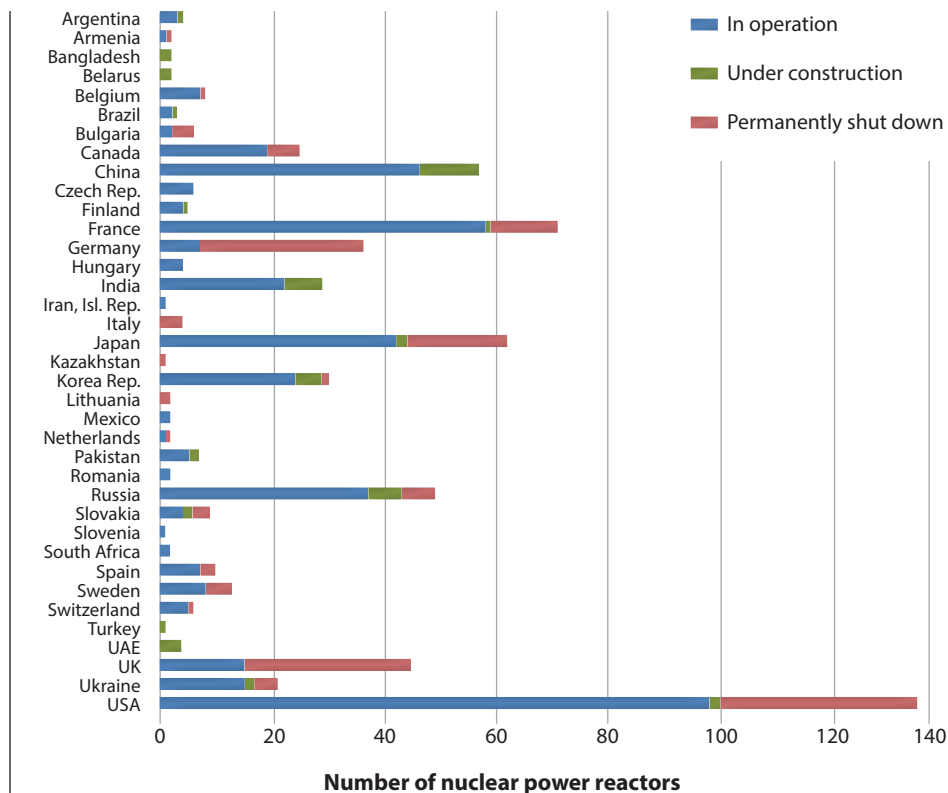
Multiple events shaped the development of the current safety and security framework. There were successes, for example, the removal of nuclear weapons from the newly independent countries of Ukraine, Belarus, and Kazakhstan and their declaration as non-nuclear weapon states. There were misses, for instance: the extended proliferation network established by A.Q. Khan of Pakistan that operated globally for years undetected.

Other events changed the public perception of the benefits and risks of nuclear technologies, and impacted policies. Accidents like Three Mile Island, Chernobyl, and Fukushima made some of the public averse to nuclear energy production, which caused governments in these countries to doubt the future of nuclear projects or abandon nuclear energy generation completely. Many developing countries need to increase their energy production in order to grow their economy and improve the well-being of their populations. The need for stable power will continue to grow in the future, especially in Southeast Asia.



Per capita energy consumption versus per-capita GDP (2011 data). The linear trend supports the notion that expanding stable power supplies is necessary in order to grow economies, and predicts that Asian countries such as China and India need to increase their energy production to improve the quality of life for their populations.

Number of nuclear power reactors by country and status, (data from IAEA, 2018). The US is a leader in nuclear power, however it has a large number of reactors permanently shut down and has very few currently under construction as public support wanes.



The US has been a leader in many nonproliferation activities and provides support to strengthen the nonproliferation regime. It is a dominant funding source for the IAEA and has initiated and implemented a number of efforts to strengthen the security of nuclear materials. However, US domestic policies have been affected by public opinion. Despite having the largest number of nuclear power reactors in the world, the US no longer seems to view nuclear power production in its future; the reactors are aging and few new nuclear reactors are being considered (see graph above). The US adopted the Nuclear Non-Proliferation Act (NNPA) in 1978 due to concerns of uncontrolled sales of nuclear fuel cycle technologies and ongoing efforts to use plutonium in civilian nuclear programs. Since adoption, this act placed limitations on domestic research and development and on international trade. Over the years there has also been a decline in the numbers of nuclear scientists and engineers, and associated research programs in the US.

Nonproliferation: Historical efforts

In the late 1970s, aligned with the direction of US, if not global nuclear efforts, the Reduced Enrichment for Research and Test Reactors (RERTR) program began. The now-defunct RERTR program enjoyed moderate success and was supported by strong policies within the US government. The most significant being the Nuclear Regulatory Commission (NRC) regulation for US research and test reactors that stipulates that if a low-enriched uranium fuel (LEU; where the ²³⁵U isotopic content is less than 25%) and funding is available then the reactor must convert to LEU fuel.

The US was also an exporter of uranium for use in both foreign research reactors as well as for medical isotope production. The "Schumer Amendment" to the Energy Policy Act of 1992 specifies additional conditions that must be met before highly-enriched uranium (HEU) can be exported from the United States. The USSR had similar uranium export programs for the supply as well as similar efforts to

convert those reactors to a lower enrichment of uranium, specifically 36%. It should be noted that the IAEA specifies that uranium with an isotopic enrichment of 20% or higher be categorized as HEU, and that there are more stringent safeguards and security requirements applicable for a state to possess that material.

The RERTR program staff would provide technical support to facilities interested in obtaining regulatory approval for conversion to LEU fuels. The RERTR program would also work to develop and test advanced replacement LEU fuels that could be used in the conversion of process. In the 1990s, the RERTR program began to experience technical difficulties with their latest high-density LEU fuel development program, and furthermore, the program seemed to lose financial support of the government, making it difficult to fully support the LEU conversion process. The RERTR program was also experiencing a significant amount of resistance from the medical isotope production community regarding conversion to LEU material, primarily based on arguments that conversion to LEU target material would be costly to refurbish the production lines, inefficient due to lower ^{235}U content, and would have an unknown regulatory approval process.

The terrorist attack on September 11, 2001, significantly changed the landscape for nuclear material threat reduction and clearly demonstrated the risk of nuclear material in civilian commerce as well as the risk of nonproliferation from non-state entities. The RERTR program was merged into a group of complimentary nuclear and radiological threat reduction programs coordinated out of Department of Energy (DOE) and National Nuclear Security Administration (NNSA) headquarters and was known as the Global Threat Reduction Initiative (GTRI). The co-location of the programs, the political attention and subsequent funding dramatically increased the rate of conversion of civilian research reactors, both domestically as well as internationally. One of the first actions that the GTRI program implemented was a conversion program of all of the remaining US HEU-fueled research reactors that had an LEU fuel available. This effort had two main purposes: to remove HEU from civilian use, and to demonstrate the commitment and leadership to all other countries that used, possessed, or supplied HEU fuel for civilian commerce.

Recent achievements

Two efforts from the GTRI that continue today and deserve mention here in part due to their broad societal impact, as well as continued relevance to nonproliferation and nuclear threat reduction priorities. The first is what is known as the “Mo-99” program, so named for the parent isotope that produces technetium-99, which is the workhorse of the nuclear medicine industry and is used globally in approximately 100,000 procedures daily. The second is the Miniature Neutron Source Reactor (MNSR) conversion program that provides a forum via the IAEA for dialogue and discussion among the participant countries (China, Ghana, Iran, Nigeria, Pakistan, and Syria) of the programmatic issues for the regulatory approval of the conversion of their respective MNSR, the procurement of LEU fuel, and manufacture of the replacement core, as well as the disposition of the spent HEU core originally in the MNSR.

For the in-depth details and story of the complexities of the Mo-99 program, the interested reader is directed first to the publications by the US National Academies of Sciences, Engineering, and Medicine, and then to the annual reports published by the Organization for Economic Cooperation and Development—Nuclear Energy Agencies High Level Group/Medical Radioisotopes.

Several of the MNSR conversion program participants have been immersed in wars, United Nations (UN) violations for nuclear activities and protracted trade and sanction discussions. It can be imagined that the MNSR conversion program, implemented with UN/IAEA oversight provides an opportune forum for discussion among the parties. Even with the difficulties facing this group, the MNSR conversion program has produced several significant accomplishments. The MNSR-IAE reactor, operated by the China Institute of Atomic Energy in Beijing, reaching criticality for the first time in 1984, was converted to LEU fuel in 2016 as a result of a cooperative project between the China Atomic Energy Authority (CAEA) and the US DOE. Ghana's Chinese-origin MNSR converted to LEU fuel in 2017, and is the first reactor of this type to be converted outside of China, establishing this cooperative effort as a model for similar cooperation on future MNSR conversions. The conversion to LEU fuel and removal of HEU fuel from Nigeria's research reactor in early 2018 resulted in all 11 research reactors in Africa being operated using LEU.

Summary

In conclusion, the United States has been a leading force in establishing and shaping the nonproliferation regime, including leadership setting up the International Atomic Energy Agency (IAEA), promoting nuclear exports control, and reducing use of nuclear weapons usable materials in civilian programs. The United States has also been a leading authority on peaceful use of nuclear energy with the world's largest fleet of nuclear power reactors. However, there has been declining support for nuclear energy in the US, resulting in fewer opportunities for education and jobs. If support for nuclear-related education, research, and industries continues to fade, it will take extra effort for the US to maintain leadership in nuclear nonproliferation.

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