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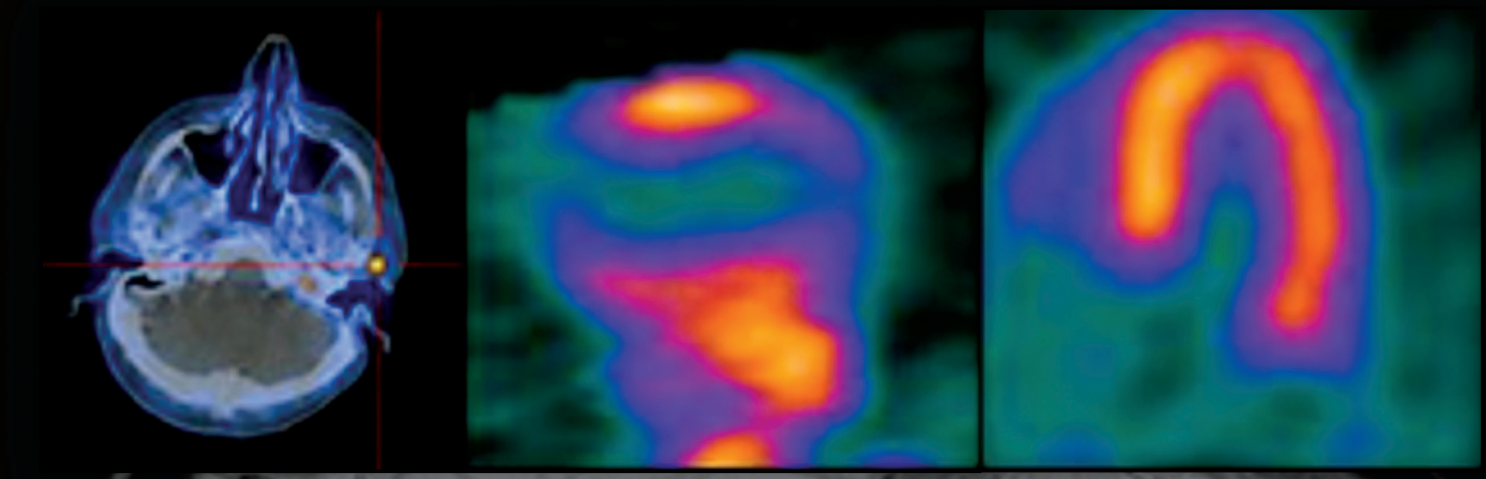

Accelerator Radioisotopes Save Lives: Part II

Los Alamos
NATIONAL LABORATORY
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Actinide Research Quarterly

Accelerator Isotopes Save Lives: Part II

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Accelerator Radioisotopes Save Lives: Part II

LANL's Role in the DOE National Isotope Program

Supplying Isotopes Worldwide for National Security, Medicine, Science, and Industry

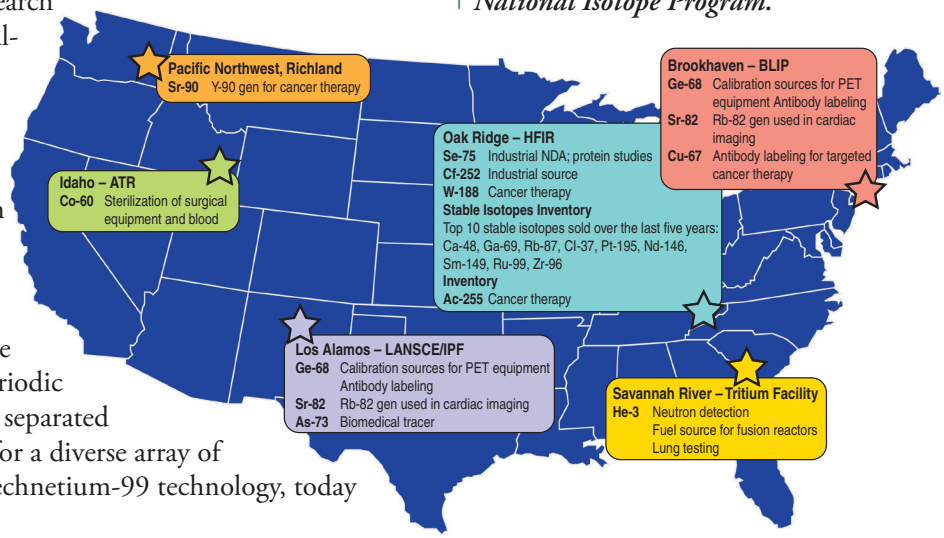
Stable and radioactive isotope products are critical components to research and applications in medicine, homeland security, environment, agriculture, and commerce. Every day, more than 40,000 medical patients receive clinical diagnostic and therapeutic procedures, many using radiopharmaceuticals (pharmaceuticals that contain radioisotopes).

Beyond the important medical and clinical applications, numerous other valuable but less well-known applications depend on isotopes as vital components. Heart imaging, cancer therapy, smoke detectors, neutron detectors, explosives detectors, environmental tracers, and oil exploration all use isotopes. Detection devices that are crucial to ensure viable national security rely on isotopes as calibration sources or as instrument components.

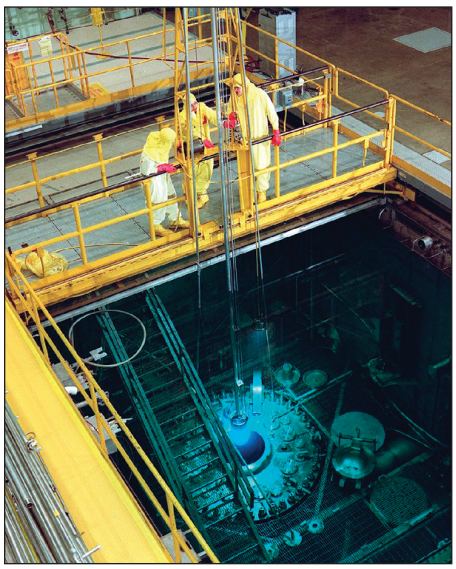
History

The Department of Energy (DOE) has been instrumental in supplying isotopes to the United States and the world, dating back to 1954 and the establishment of the Atomic Energy Act. Since then, the DOE, through its National Isotope Program, has led the government's efforts to maintain and enhance the national isotope technology infrastructure and to provide isotopes to medicine, science, and industry. Linking government investments in isotope production capabilities and development of new stable and radioactive isotopes has led to advances in medicine; basic research in physical, earth, and life sciences; agriculture; industry; defense technologies; and national security.

The availability of many of these isotopes depends on the safe and effective operation of the DOE isotope production facilities. The calutrons at Oak Ridge National Laboratory, built to separate uranium isotopes for the Manhattan Project in the 1940s, were used to separate isotopes of nearly every element in the periodic table. Until the early 1990s, these devices separated hundreds of kilograms of stable isotopes for a diverse array of applications. The first molybdenum-99/technetium-99 technology, today



Current primary production sites used by the DOE National Isotope Program.



Technicians replace a fuel element at the High-Flux Isotope Reactor at Oak Ridge National Laboratory.

Idaho National Laboratory's Advanced Test Reactor produces valuable medical isotopes, including cobalt-60.

used in about 90 percent of all nuclear medicine applications, was developed at Brookhaven National Laboratory and sponsored by the DOE in the 1950s.

Over the past several decades, emerging technologies have reduced health-care costs and improved the quality of life for innumerable patients. As an example, the development of positron emission tomography (PET) has improved the ability of physicians to diagnose illnesses. PET uses positrons emitted from radionuclides to create images of organs to diagnose cancer. Two critical isotopes for PET technology, rubidium-82 and germanium-68, are provided by the National Isotope Program. The use of rubidium-82 (half-life = 75 seconds) with PET is well established in myocardial perfusion imaging for the diagnosis of coronary artery disease. Rubidium-82's parent, strontium-82 (half-life = 25.5 days), is immobilized on an ion-exchange column, from which the rubidium-82 can be eluted right before each PET scan. Germanium-68 is used as rod source for the detector calibration for PET scanners and to measure non-uniform patient attenuation. Together, Brookhaven and Los Alamos National Laboratories have been providing these materials enabling tens of thousands of diagnostic procedures every month at U.S. hospitals.

Production challenge

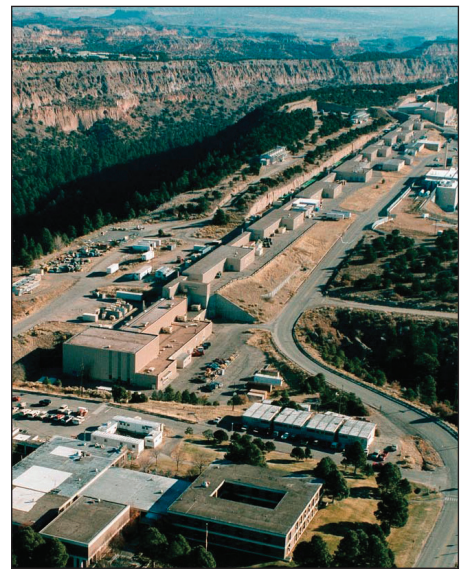
Providing a reliable supply of radioactive isotopes is challenging because of the limited number of facilities available for production and processing. To provide isotopes year-round, a network of production sites must be integrated into a robust and reliable production schedule. Cooperative agreements with domestic and foreign isotope manufacturers and research organizations to supply irradiated targets are a major component of the National Isotope Program's strategy to improve reliability of supply and ensure on-time deliveries to customers when DOE accelerators and reactors are not operating. Currently, silicon-32 is produced by irradiating potassium chloride at Canada's TRIUMF facility and then processed at Los Alamos.

The National Isotope Program produces radioisotopes mainly at three locations: Los Alamos and Brookhaven, using accelerators, and Oak Ridge, using a reactor. Each location has associated hot cells for the handling and processing of radionuclides. The Brookhaven Linac Isotope Producer (BLIP) is a linear accelerator that can deliver protons with up to 200-megaelectronvolt (MeV) energy and 145 microamperes beam current intensity. The National Isotope Program operates the BLIP at only 117 MeV, as the demand for isotope production requiring 200-MeV irradiation is very small. The BLIP operating schedule complements the operating schedule of the Isotope Production Facility (IPF) at Los Alamos. The IPF produces radioisotopes by using a 100-MeV proton beam obtained by diverting a portion of the main Los Alamos Neutron Science Center (LANSCE) beam before it enters the final portion of the accelerator and directing it to a new isotope production targeting area. Both IPF and BLIP's major products include germanium-68, strontium-82, arsenic-73, zinc-65, and sodium-22.

The High-Flux Isotope Reactor (HFIR) at Oak Ridge provides one of the world's highest steady-state neutron fluxes. The HFIR contains numerous target locations within the core and the reactor beryllium reflector, with fluxes ranging from 400 trillion ($4x10^{14}$) neutrons per square centimeter per second (cm^2sec) to $3x10^{15}$ neutrons/cm^2sec, which are ideally configured for the production of the program's radioisotopes. In addition, three centrally located hydraulic tubes are also available for the rapid insertion and removal of targets requiring short irradiation times. A wide variety of radioisotopes are currently produced at the HFIR, including tungsten-188, nickel-63, californium-252, selenium-75, and lutetium-177. HFIR has historically produced more than 100 additional radioisotopes, many of which are now commercially available.

The Advanced Test Reactor (ATR) at Idaho National Laboratory has been used to produce high- and low-specific activity cobalt-60. It provides a maximum flux of about $2–4x10^{14}$ neutrons/cm^2sec. Recently, the state of Idaho has agreed to finance the installation of a hydraulic tube at the ATR. This upgrade will promote cooperative opportunities for Idaho National Laboratory and provide additional reactor target volume. At Pacific Northwest National Laboratory, hot-cell facilities are used to purify and distribute strontium-90, and helium-3 is distributed from the Savannah River Site.

At each facility, the radioisotope production mission shares the reactor or accelerator with other ongoing basic-energy sciences or defense missions that are generally much larger and so control facility schedules and priorities. Because radioisotope production is often not the first priority, it becomes dependent on the operating constraints of the larger, primary missions. For these reasons, the National Isotope Program needs to continue importing irradiated targets or processed isotope products from foreign producers to complement production capabilities and to improve isotope availability within the United States.



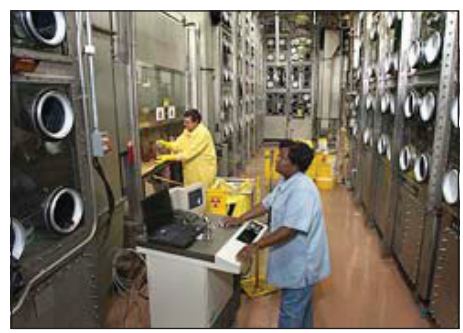
The Isotope Production Facility at the Los Alamos Neutron Science Center uses a 100-megaelectronvolt proton beam to produce a variety of radioisotopes.

Radioisotope production at Los Alamos

Los Alamos has a long history of producing radioisotopes for applications in medicine, biology, nuclear physics, and national security. From 1974 to 1998, isotopes were produced at Area A at the end of the beam line at what was then called the Los Alamos Meson Physics Facility (LAMPF) and is now called LANSCE. The isotopes were produced via spallation using the available 800-MeV proton beam.

Spallation is a nuclear process in which a heavy nucleus emits a large number of nucleons as a result of being hit by a high-energy particle, thus greatly reducing its atomic weight. Target materials like rubidium bromide were irradiated to produce a variety of lighter isotopes like silicon-32 (an environmental diagnostic), copper-67 (used for treatment of non-Hodgkins Lymphoma), and germanium-68 (used for instrument calibration).

During the period from 1998 to 2004, isotopes were produced from target materials irradiated at other accelerator or high-energy cyclotron facilities because of a disruption in the proton beam at LANCSE. Some of these facilities



Savannah River Site employees work in the Tritium Facility.

Brookhaven's 200-megaelectronvolt linear accelerator provides protons for use at the Brookhaven Linac Isotope Producer.

Researchers at Pacific Northwest National Laboratory have developed a process to make ultrapure yttrium-90, an important material for cancer therapy.

included Russia's Institute of Nuclear Research (INR), South Africa's iThemba Laboratory for Accelerator Based Sciences, and Canada's TRI-University Meson Facility (TRIUMF). Target materials were received at Los Alamos's TA-48 Hot Cell Facility and purified for domestic distribution during this period.

The Los Alamos collaboration with Russia's INR was facilitated through the NNSA's Global Initiatives for Proliferation Prevention office, which focused on providing former Soviet Union weapons scientists with peaceful applications of their knowledge of nuclear physics. INR was instrumental in supplying irradiated rubidium-metal targets for the production of strontium-82. At that time, strontium-82 was seen as an emerging isotope for cardiac imaging. This relationship continues today with INR supplying roughly 8 percent of the U.S. supply of this critical isotope.

The mission for the accelerator facility changed in the 1990s and enabled the Los Alamos radioisotope program to upgrade its capabilities. Los Alamos isotopes are now produced at the 100-MeV IPF at LANSCE. Funded by the DOE Office of Nuclear Energy, the IPF was commissioned in 2003 at the cost of $24 million. It has been instrumental in providing isotopes to domestic and international customers like GE Healthcare, the Environmental Protection Agency, and a variety of research institutions.

Safely transporting isotopes

The National Isotope Program's products and services are provided to hundreds of customers worldwide. Each year more than 450 shipments of stable and radioactive isotopes serve customers in science, hospitals, and commerce. Shipments are governed by Department of Transportation and Nuclear Regulatory Commission regulations to ensure public safety and to protect the environment.

The mode by which radioactive materials are shipped depends on the size and weight of the packages, decay properties and quantities of the radioactive material, and the availability of and distance between transportation facilities. Small quantities are usually transported by air, while larger quantities are transported by highway.

The National Isotope Program continues to ensure an adequate fleet of shipping casks to carry a variety of product radioisotopes. The program either owns or uses 17 different kinds of Type A containers and six different kinds of Type B containers. Most of the containers are reusable. The most common non-reusable Type A containers can be purchased commercially whenever the supply is depleted.

New name, expanded mission

In fiscal year 2009, the National Isotope Program was transferred from the Office of Nuclear Energy to the Office of Nuclear Physics in the Office of Science. The mission of the renamed Isotope Development and Production for Research and Applications Program is to develop, produce, and distribute

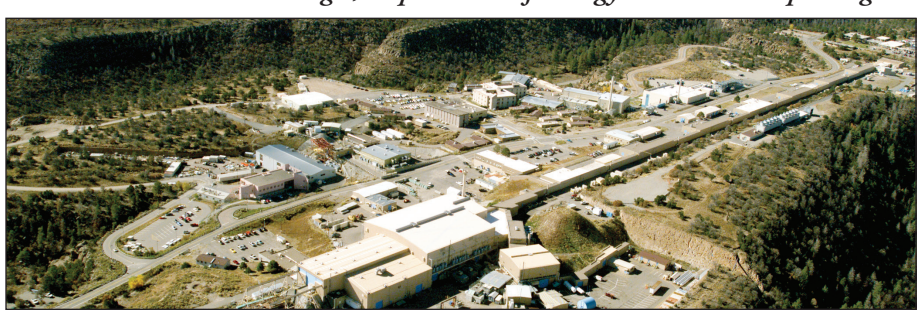
stable and radioactive isotope products that are in short supply. The program is supporting innovative research and development to significantly strengthen its capabilities to produce stable and radioactive isotopes and intends to increase its coordination and support of isotope production at a larger suite of universities, national laboratories, and commercial accelerator and reactor facilities. These efforts are targeted to promote a reliable supply of domestic isotopes and ensure a reliable supply of isotopes for research applications.

Los Alamos has been provided the opportunity to contribute to the program using its capabilities and expertise in isotope production. The latest federal budget proposal does not include funding to continue necessary upgrade work on the aging Los Alamos accelerator, a project known as LANSCE-R. If funding isn't secured, not only will LANSCE not be available for work critical to the stockpile stewardship program, the future supply of life-saving isotopes will be greatly threatened. Currently, Los Alamos staff are evaluating options to produce actinium-225, a promising candidate for alpha-immunotherapy, at the IPF. Experiments to measure cross sections for a range of proton energies (100, 200, and 800 MeV) have been initiated, and small amounts of actinium-225 have been chemically separated and analyzed.

Partnering with universities, such as the University of Missouri and UC Davis, will enable Los Alamos to contribute to educating students and training the next generation of nuclear scientists. Given the unique capabilities at the IPF and the future prospects and the opportunities at the Materials Test Station, Los Alamos is posed to play a critical role in the National Isotope Program and continues to be essential to the program's mission to reliably supply isotopes for a wide array of applications.

The Los Alamos Isotope Program has established a strong tradition of providing quality isotope products and services for the medical, industrial, and R&D sectors over its 35-year history. More than 250 customers have made use of the program. This issue of *Actinide Research Quarterly* is a follow-on to an article published in *Los Alamos Science* (No. 30, 2006), which can be read online at http://la-science.lanl.gov/index.shtml. The following articles provide a description of some exciting, isotope-related science and technology areas under development at Los Alamos National Laboratory.

—Wolfgang Runde
Manager, Department of Energy National Isotope Program

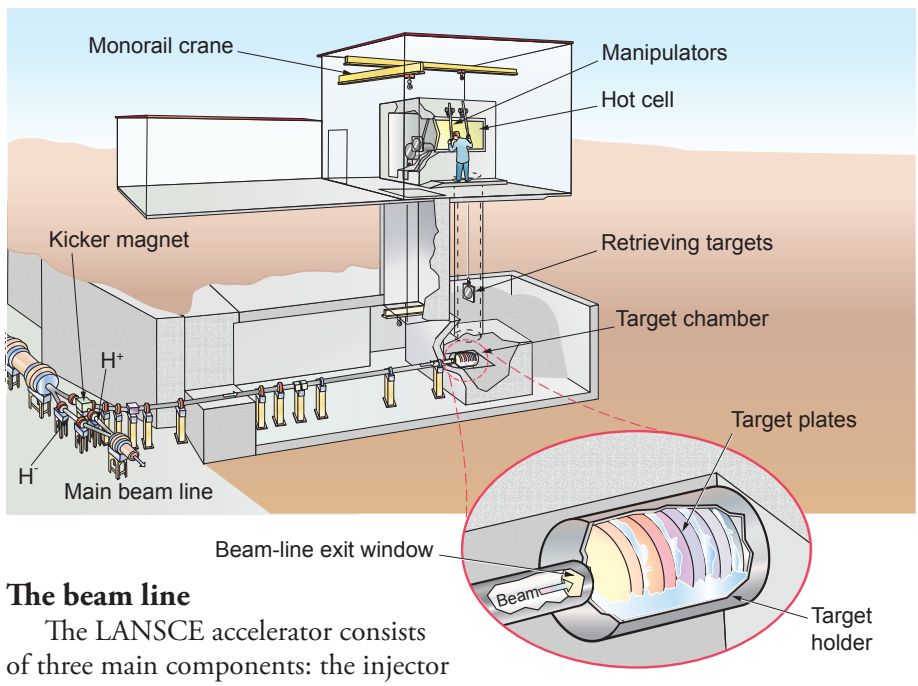


The Los Alamos Neutron Science Center and its half-mile-long accelerator provide physicists worldwide with a unique user resource.

The Isotope Production Facility at TA-53

This article was contributed by Meiring Nortier and Hong Bach of the Inorganic, Isotope and Actinide Chemistry Group.

The Isotope Production Facility (IPF) is a state-of-the-art facility located near the front end of the Los Alamos Neutron Science Center (LANSCE) accelerator. It uses high-intensity protons with a nominal energy of 100 million electron-volts (MeV) to irradiate materials for radioisotope production. It consists of two levels: the lower level houses the beam line, target chamber, and target cooling system; the upper level houses the equipment room and one hot cell. The IPF has provided Los Alamos with the capability to produce isotopes on a larger scale in a more-efficient process and is positioning the Laboratory globally as a unique, dedicated isotope-production site.



Cross section of the Isotope Production Facility.

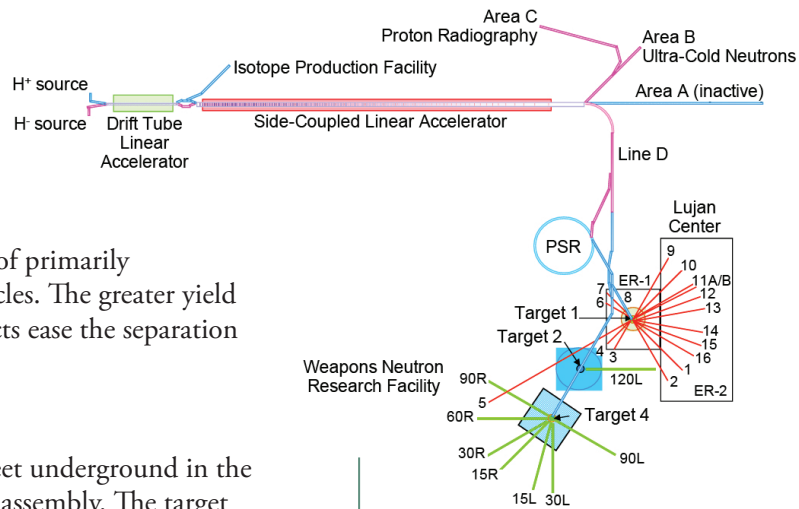
The beam line

The LANSCE accelerator consists of three main components: the injector accelerating protons up to 750 kiloelectronvolts (keV), a drift-tube linac for acceleration up to 100 MeV, and a side-coupled cavity linac to further accelerate the protons to 800 MeV. The new beam line for IPF diverges at the transition between the 100-MeV drift-tube linac and the 800-MeV side-coupled linac, where 100-MeV protons are extracted and magnetically deflected to the IPF. The 100-MeV IPF beam line ends at the target chamber, where the beam passes through an exit window and irradiates a specially designed multiple-target assembly.

The nuclear reactions necessary to produce new isotopes are quite different at 800 and 100 MeV. Bombardment of material with high-energy 800-MeV protons initiates a nuclear reaction called spallation in which the bombarded target nucleus breaks into smaller fragments or ejects fragments of the nucleus. The spallation of a nucleus leaves behind a wide array of isotopes, creating a challenge for the chemists to separate generally small quantities of a desired

isotope from a number of unwanted impurities. This separation is time-consuming and creates a large volume of radioactive waste that has to be disposed of properly.

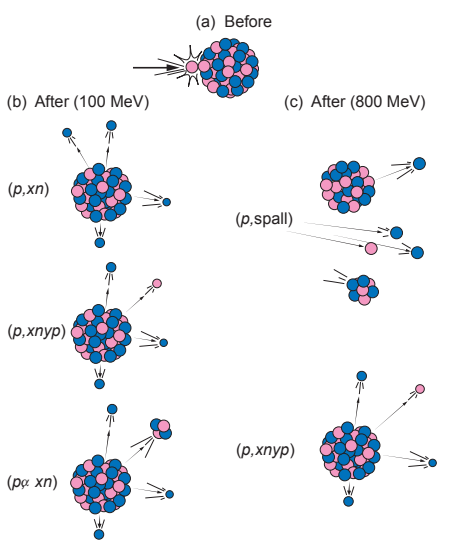
Irradiation with the medium-energy 100-MeV protons increases the yields of the desired isotopes and reduces the fraction of unwanted byproducts. In general, the 100-MeV protons induce a release of primarily neutrons and occasionally protons and alpha particles. The greater yield of desired products and lesser amount of byproducts ease the separation task significantly.



Schematic of the Los Alamos Neutron Science Center.

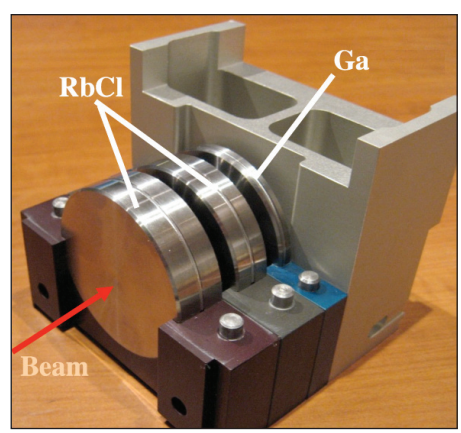
Target irradiation

The target irradiation at IPF occurs about 40 feet underground in the water-cooled target chamber containing the target assembly. The target assembly consists of a stack of target discs arranged in a holder along a horizontal center line of the incoming proton beam. The targets are separated by about 5 millimeters (mm) to allow flowing cooling water to remove the intense heat deposited by the proton beam. The stacking of up to three targets allows one to vary the energy of the protons impinging on each target as the protons lose energy along their path through the target materials.

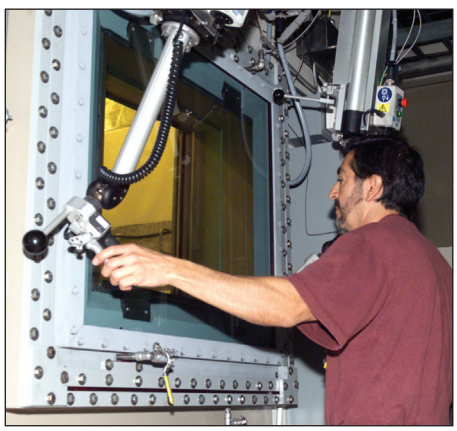


Proton-induced nuclear reactions at 800 and 100 megaelectronvolts (MeV). At energies of up to 100 MeV, protons cause the release of one or several neutrons and sometimes the release of a proton or an alpha particle in addition to neutrons. At 800 MeV, protons include spallation reactions that break apart the nucleus into chunks or cause large numbers of neutrons and smaller numbers of protons to evaporate from the nucleus.

The front target experiences close to full-energy 100-MeV protons, while the next two targets typically experience incident proton energies of 65 and 30 MeV, respectively. Residual low-energy protons exiting the last target in the stack are stopped at an aluminum block. The target configuration is designed to maximize production yields and to allow the simultaneous production of different isotopes at different beam energies, using different target materials. The irradiated targets are retrieved through the



A target holder with two rubidium-chloride (RbCl) targets in the 90-megaelectronvolt (MeV) and 65-MeV energy slots, and one gallium (Ga) target in the 35-MeV energy slot.



The Isotope Production Facility hot cell with manipulators and leaded glass window provides safe handling of highly activated materials.

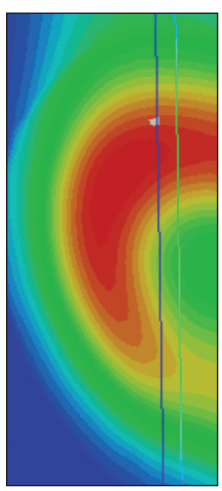
vertical shaft using a remotely operated target-transfer system. This system guides the target stack between the target chamber and the hot cell and allows operators to load and remove targets without entering the beam tunnel.

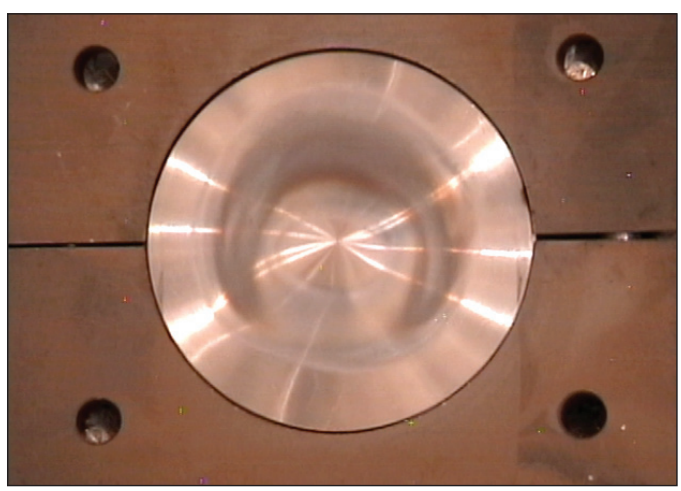
The hot cell and remote target-handling systems provide a shielded working area where fresh targets are remotely placed into the target holder and irradiated targets are disassembled and loaded into shipping casks. The hot cell is shielded on three sides and on the top with concrete and steel, while a shield door in the rear wall provides access to the interior of the hot cell for maintenance. Irradiated targets containing highly radioactive isotopes are loaded into a certified and heavily shielded cask and shipped from the IPF to the chemical processing facility at TA-48. An overhead monorail crane transfers the cask to and from the truck for transportation between facilities.

The targets irradiated at IPF have a nominal diameter of 50 mm and typically consist of a durable metal capsule filled with metal or salt. The capsule halves are machined from metals such as 316 stainless steel, inconel 625, or niobium with a window thickness of 0.3 mm. For strontium-82 production, rubidium-chloride salt pucks are cast and then sealed inside an inconel capsule under vacuum by electron beam welding. For the production of germanium-68, gallium metal is encapsulated in niobium. Prewelded niobium capsules are filled with liquid gallium through a fill port, which is plugged and sealed under vacuum by electron beam welding.

Thermal analyses of rubidium-chloride targets under bombardment at a maximum beam current of 250 microamps (μA) predict a peak temperature in the target of close to, but not exceeding, the boiling point of the rubidium-chloride salt. By contrast, the peak temperature in a gallium target at 250 μA does not challenge the material boiling point at all. This is due to a higher thermal conductivity and a higher boiling point of the gallium over that of rubidium-chloride salt.

Comparison of an expected temperature distribution pattern (left) with an actual target discoloration pattern on the face of a gallium target.

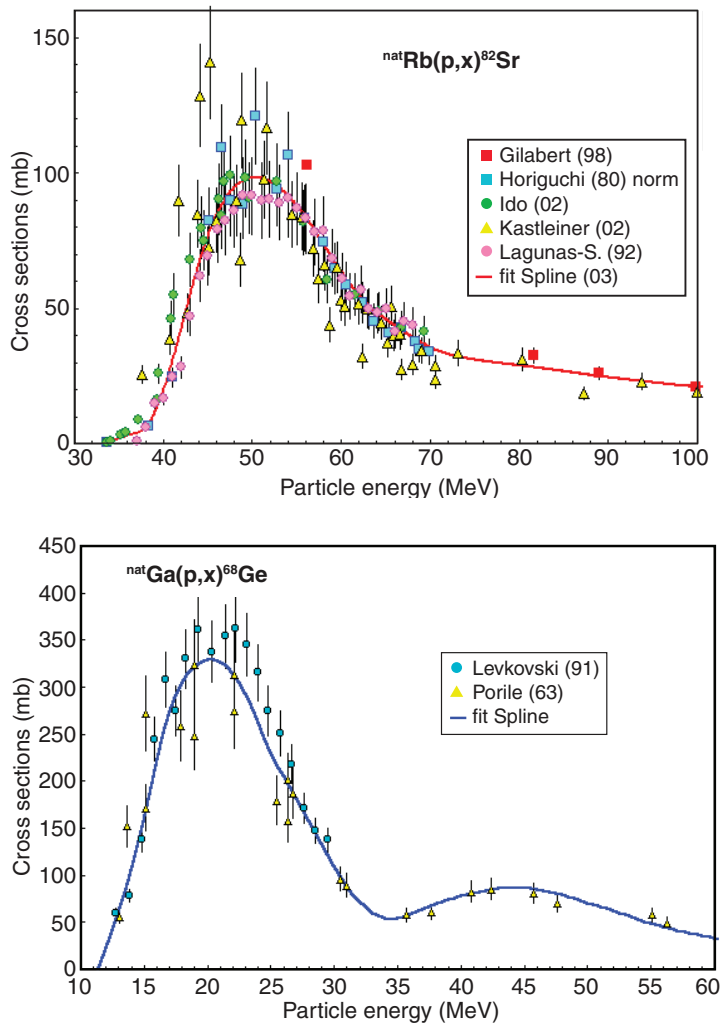




While an accurate prediction of the effects from all the thermal processes occurring in a fully or partially molten target is difficult, a comparison of predicted temperature distribution patterns with corresponding discoloration patterns on the faces of actual irradiated targets many times show very good agreement. Such positive visual feedback provides increased confidence in the predictability of our models describing all the coupled mechanisms that participate in establishing the steady-state temperature distribution in a partially molten target under bombardment.

Target development

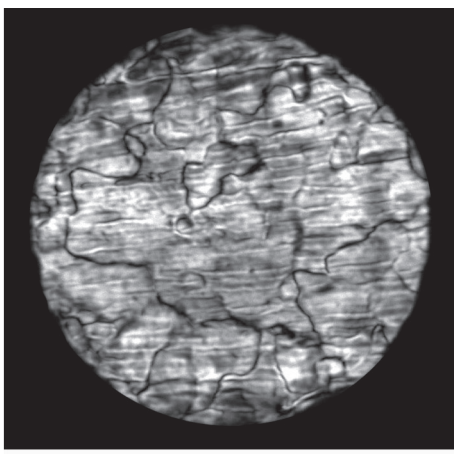
The targets are positioned within the stack to maximize the rate at which the desired isotope will be produced. For example, a rubidium target, which is typically in the form of a chloride and is used to make strontium-82, is placed in the middle of the stack because the cross section for that production reaction is greatest for incident protons with energies between 45 and 65 MeV. Similarly, a gallium target is placed at the rear of the stack to use the energy range below 30 MeV for the production of germanium-68. Isotope production rates for each target are predicted from these excitation functions and depend on the position in the stack, the reaction cross section, and the target thickness. Irradiation times are determined by those production-rate calculations and the amount of material that must be produced to satisfy customer requirements for the isotope.

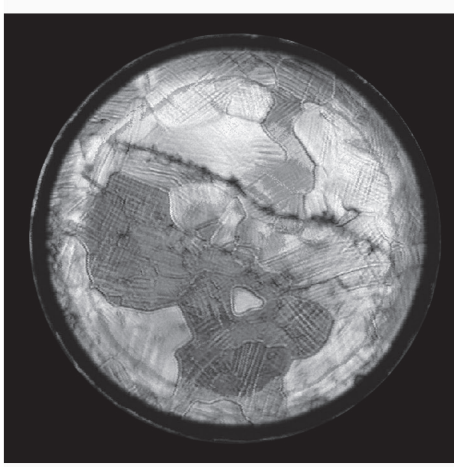


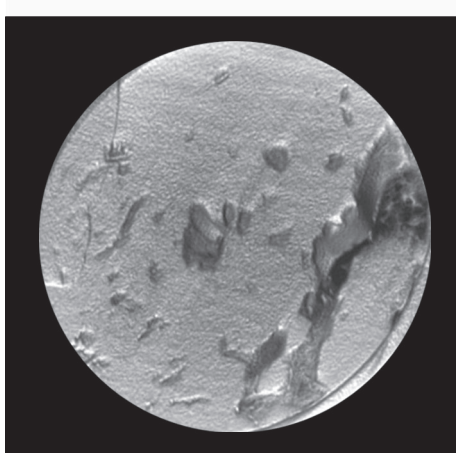
Excitation functions for the production of strontium-82 (top) and for the production of germanium-68 (bottom).

Because the proton beam deposits up to 8 kilowatts (kW) of power into a single target and its capsule during irradiation, the target is immersed in fast-flowing cooling water. For target materials that require encapsulation, the target capsules are exposed for extended periods to intense radiation, thermally induced stress, and chemicals. The structural integrity of the target capsules is of crucial importance for containing both produced radioisotopes and target material. In the case of gallium metal, which is used as the target material for the production of germanium-68, the niobium capsule must be sufficiently robust to remain leak tight while subjected to intense thermal stress cycles and a high-radiation environment. Yet the capsule window must be sufficiently thin so that the proton beam is not significantly attenuated by it.

Target capsule failure not only increases radioactive waste, cleanup and maintenance costs, and the risk of workers' exposure to radiation, but also lowers product yields, revenue, and the reliability of the isotope supply. It is imperative







that the integrity of the targets is verified before insertion in the beam line to prevent target failures, but it is also sometimes necessary to inspect targets periodically during irradiation for capsule deterioration. Digital radiographic and ultrasonic images can be used for this purpose. Computed tomography and digital radiography results can also be used to determine void content and distribution of gallium in the targets.

Efforts to incorporate these imaging techniques into the IPF hot cell are under way. Researchers are studying the effects of irradiation damage along with the simultaneous effects of thermal, hydraulic, chemical, and mechanical processes on the structural integrity in the irradiation environment. The capability of nondestructive analysis of irradiated targets will greatly enhance the ability to prevent costly target failures.

Future isotope portfolio

The vast majority of available beam at IPF is presently used for the production of commercial isotopes. Strontium-82, used in cardiac imaging to diagnose blood-flow diseases, and germanium-68, used to calibrate the hundreds of PET scanners in the United States, are the main isotopes produced. Other research isotopes include arsenic-73, used as tracer in environmental studies; sodium-22, applied as a positron source; and several isotopes for applications related to homeland security.

With the recent changing mission of the DOE Isotope Program within the Office of Science, the emphasis is shifting to re-establishing the production of isotopes needed by the research community. The Los Alamos team is currently investigating the feasibility to produce several short-lived isotopes at IPF. Examples are actinium-225, an alpha-emitting isotope desired for radio-immunotherapy studies, and yttrium-86 and copper-67, both promising candidates in cancer detection and treatment. The challenge in producing such isotopes is in the fast isotope separation and transportation to the customers to minimize loss due to radioactive decay.

Above left: Ultrasonic scans taken on a 5.17-millimeter-thick niobium starting rod (top), a gallium-filled capsule showing niobium grains and gallium wetting of the niobium window (center), and a computed tomography image of the void distribution of the gallium in a partially filled niobium capsule (bottom).

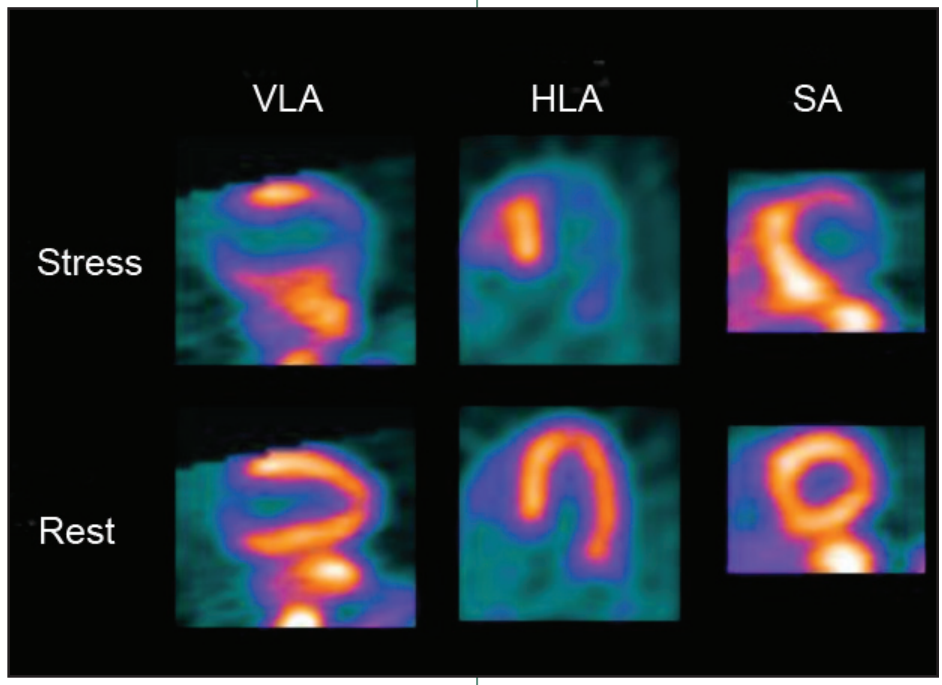
Radioisotopes for Medical Applications

This article was contributed by Kevin John of the Office of Science Programs.

Two key radioisotopes for medical applications are currently being produced at the Isotope Production Facility (IPF): germanium-68 (half-life = 271 days), used for calibrating positron emission tomography (PET) scanners, and strontium-82 (half-life = 25.55 days), the parent of the very short-lived rubidium-82, used for PET scans of the heart. The strontium-82 product is provided as an aqueous solution for use in what is called an isotope generator.

The generator technology immobilizes the strontium-82 on a metal-oxide support allowing elution of the daughter isotope rubidium-82 (half-life = 75 seconds), which is injected into patients as a saline solution (30–60 millicuries per patient dose). Rubidium-82 follows the potassium pathway in the body and collects in heart tissue prior to positron emission to form stable krypton.

PET technology takes advantage of a unique aspect of positron emission in an electron-rich environment such as the human heart. Specifically, the positron emission is quickly followed by an annihilation event after the positron has traveled up to a few millimeters. The positron–electron annihilation process provides two completely orthogonal gamma rays at 511 kiloelectronvolts (keV).

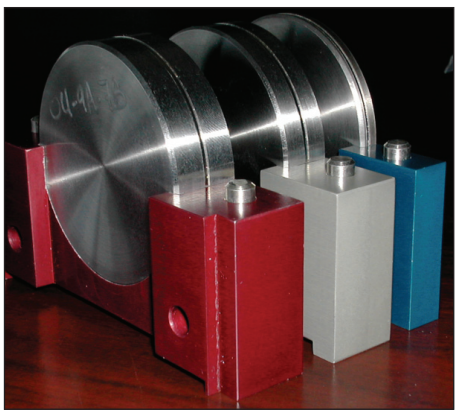


Rubidium-82 positron emission tomography (PET) stress–rest images.

The orthogonal gamma emission is a direct result of conservation of angular momentum associated with the positron–electron annihilation process. This allows a PET scanner to triangulate where the positron emission occurred, making it possible to generate an image of the heart. Damaged areas of the heart collect less rubidium-82 because of a lack of blood flow and therefore appear darker in the PET images, allowing early diagnosis of a variety of cardiac conditions.

Germanium-68 is an integral part of the PET imaging process as its long half-life allows it to be manufactured in well-defined objects called "phantoms" that are essential for the quality-assurance process for PET machines (there are roughly 1200 PET centers worldwide.

The impact on U.S. healthcare for these two isotopes is significant, given the fact that strontium-82 is used for roughly 10,000 cardiac-imaging procedures each month. Furthermore, germanium-68 phantoms and line sources are used to calibrate PET instruments that are used in millions of medical imaging procedures every year. In addition, germanium-68-containing line sources are used to measure patient-specific attenuation factors that provide a background correction for a PET scan.



A target stack for strontium-82 and germanium-68 production.

Production of strontium-82 and germanium-68 at IPF

The IPF receives its proton beam from the transition region between LANSCE's 201 and 805 linear accelerators (linacs). The beam is bent from the LANSCE axis using a powerful set of magnets to divert beam to our production facility, where it is typically divided over three water-cooled target slots. As beam passes through each sequential target and cooling-water channels, the energy is reduced with energy ranges of 93–70 (high), 65–40 (medium), and 31–0 (low) MeV. None of the beam is wasted during production.

Strontium-82 is formed at IPF in the high- and medium-energy target slots by irradiating two targets consisting of a rubidium–chloride puck clad in a nickel-based superalloy. The rubidium–chloride target is heated by the proton beam such that it becomes molten in the target capsule even though the targets are cooled by a continuous flow of water. Targets are kept in beam twenty-four hours a day for an average of eight to ten days during a typical irradiation cycle. During irradiation, rubidium atoms in the target material undergo (p,4n) and (p,6n) nuclear reactions where the incident proton (the "p") ejects four to six neutrons (the "n") from the target nucleus.

This is a very low-probability event with only a small percentage of the protons engaging in nuclear reactions; the majority of the incident protons react with the electrons in the target matrix resulting in the formation of hydrogen atoms. The associated chlorine atoms undergo similar reactions in the target material and the resultant products must be separated during subsequent chemical processing.

Cleaner product is available if a pure rubidium-metal target is irradiated and higher yield is possible due to the increased rubidium atom density; however, rubidium metal is extremely pyrophoric and reacts violently with water. For this reason, IPF has avoided pure rubidium-metal target irradiations, but Russia's Institute of Nuclear Research (INR) and South Africa's iThemba Laboratory for Accelerator Based Sciences produce strontium-82 using this reactive target material. Germanium-68 is typically produced in the low-energy target slot using liquid gallium targets encapsulated in a niobium shell. Germanium-68 is produced via (p,2n) and (p,4n) nuclear reactions.

Once targets have been irradiated, they are transported to the TA-48, RC-1, Hot Cell Facility using a specially configured Department of Transportation Type B container. This production process continues for strontium-82, germanium-68, and R&D isotope production during the approximately six-month operational window at LANSCE.

Chemical processing operations

The TA-48 hot cells consist of a connected bank of thirteen hot cells with a specialized function for each cell. The cells are each configured with 20 inches of ferro–phosphorous concrete to provide adequate shielding from the irradiated IPF targets and subsequent isotope products. The hot-cell windows consist of five panes of successively thicker-leaded-glass, with a specialized oil

separating each window that is optically matched to the glass permitting a clear, undistorted view of the hot-cell interior.

Cells are connected to one another by a train that can be used to transport targets, chemicals, and equipment needed for a given operation. The facility is certified by the Federal Drug Administration, given that some of the materials produced there are deemed as pharmaceutical ingredients for end use in humans.

Targets are introduced into the hot cell where they undergo several processes. They are first punched to provide access to the target materials. This step is followed by target dissolution that usually involves an aqueous acidic solution, depending on the target material.

In the case of rubidium-metal targets, a hot cell must be purged with argon prior to sequential dissolution of the target material in isopropanol and water to control reactivity in this process. A variety of conventional wet-chemistry techniques are then used in the hot cell—a set of tasks requiring operators to take years of training in the art of remote manipulation—to isolate the isotope of interest.

Final products undergo rigorous radioassay and analysis for stable isotopes to ensure that the product is free of impurities that may preclude the intended end use. Products are typically packaged in DOT Type A containers that can be shipped around the country and the world. The program has historically made approximately 140 shipments each year to domestic and foreign end users.

Irradiated targets are transported to TA-48 for chemical processing within a bank of hot cells that are shielded by 20 inches of concrete.

Looking ahead

Strontium-82 production has become all the more critical because of the outages of key reactors in Canada because of technical issues with the Chalk River Reactor and the Multipurpose Applied Physics Lattice Experiment (MAPLE) reactors. These reactors are used in the production of roughly 50 percent of the world's supply of molybdenum-99, the parent isotope of technetium-99m (m = metastable). Technetium-99m is an isotope used in about 20 million medical procedures yearly; 60 percent of which are used in the areas of cardiology and oncology.

Widespread use of strontium-82/rubidium-82 generators as a replacement for their molybdenum-99/technetium-99m analogs requires a paradigm shift in the clinical-imaging arena. Technetium-99m decays via gamma emission and is used with conventional single-photon emission-computed tomography (SPECT), and rubidium-82 decays via positron emission, which is detected via less conventional (i.e., expensive) PET machines. However, use of PET technology is growing at a rate of approximately 20 percent each year.

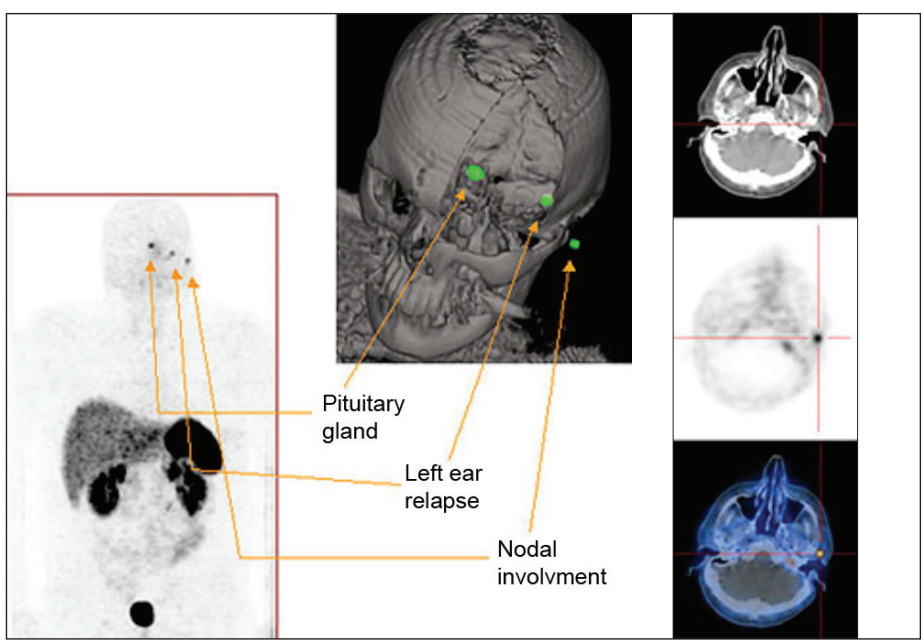
In addition to strontium-82, the daughter isotope of germanium-68–gallium-68 (half-life = 68 minutes) is beginning to receive attention in the clinical imaging arena. Gallium-68 readily forms stable complexes with DOTA (a synthetically flexible metal chelating agent), allowing peptides and other small molecules to be radiolabeled at high specific activities. Gallium-68

has a positron yield as high as 89 percent, making it well suited for PET imaging. Its half-life of 68 minutes is compatible with a variety of radiopharmacy methods of interest in diagnostic imaging and therapy.

The 271-day half-life of the parent isotope germanium-68 allows the use of the germanium-68/gallium-68 generator for at least one year in locations that do not have access to an accelerator or cyclotron. The use of germanium-68/gallium-68 generator technology is beginning to lead to widespread acceptance of this isotope in some of the areas where molybdenum-99/technetium-99m generators are conventionally applied.

Strontium-82 and germanium-68 aren't the only isotopes of interest for Los Alamos' Isotope Program. The program has received funding to pursue the irradiation of natural-abundance thorium targets for the production of actinium-225, an isotope that shows great promise in clinical applications for alpha immunotherapy. Actinium-225 decays via four alpha particles to create bismuth-209; the resultant alpha-particle cascade provides a potent mechanism to destroy tumor sites while leaving surrounding healthy tissue relatively unperturbed.

In addition to thorium-target irradiations, IPF is interested in the irradiation of other actinide target materials that will provide unique isotopes like neptunium-236 that have national security implications.



A positron emission tomography-computer scan of a neuroendocrine tumor using a gallium-68–DOTA complex. The central scan of the cranium by Policlinico S. Orsola-Università di Bologna in Italy was named Society of Nuclear Medicine Image of the Year 2008.

http://web-cwcamer.gehealthcare.com

Radionuclide Generators: Portable Sources of Short-Lived Medical Isotopes

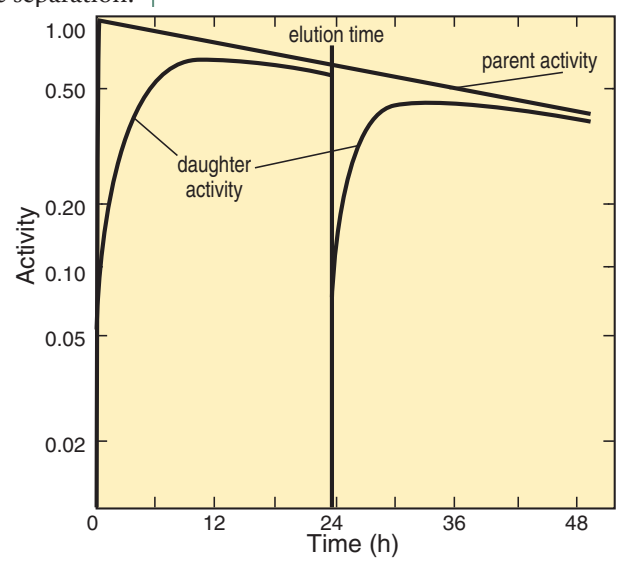
This article was contributed by Michael Fassbender of the Inorganic, Isotope and Actinide Chemistry Group and Kevin John of the Office of Science Programs.

A radionuclide generator is a pair of radioactive nuclides where one (the "daughter") is the decay product of the other (the "parent"). The daughter is typically a rather short-lived isotope with either imaging or therapeutic properties. The advantage of such a generator is obvious: it enables the on-site supply of a radioisotope that could otherwise not be provided due to the lack of a more elaborate production facility (i.e., a reactor or cyclotron) in close proximity.

Typically, the radionuclide generator functions on the basis of different chemical or physisorption behavior of parent and daughter. While the parent remains contained in one phase, the daughter is released from the same phase and moves into a second phase. Thus, a separation of both nuclides is achieved after the daughter isotope has accumulated to the desired activity level. Typical techniques used are chromatographic absorption, distillation, or phase separation.

Frequently, the half-life of the parent is much longer than the daughter's. After daughter-isotope removal (i.e., after "milking" of the generator), the daughter activity buildup or "recharge" process is governed by both the initial parent activity load and the half-life of the daughter. One may arbitrarily define a "quasi-equilibrium" after the elapse of ten daughter half-lives, after which the generated daughter activity has built up to saturation, so that another batch can be removed from the generator. The "shelf-life" of the generator, on the other hand, is dictated by the parent's half-life.

In a generator where the half-life of the parent nuclide is much longer than that of the daughter nuclide, 50 percent of equilibrium activity is reached within one daughter half-life and 75 percent within two daughter half-lives. Hence, milking the daughter nuclide off the generator is reasonably done after at least one daughter half-life.

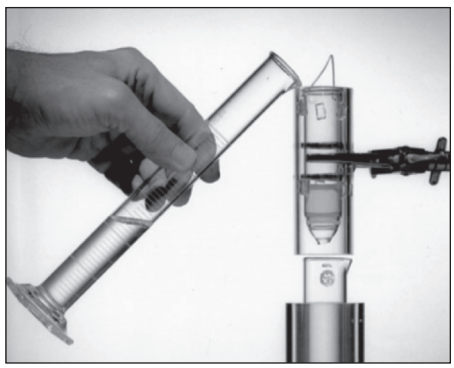


Parent and daughter activity curve in a regularly milked radionuclide generator.

Molybdenum-99/technetium-99m radionuclide generators

The most widely used radionuclide generator system is the molybdenum-99/technetium generator on which much of current routine nuclear imaging relies. Technetium-99m (m = metastable) is used in single-photon-emission computed tomography (SPECT). Worldwide, technetium-99m is used in about 20 million nuclear-medicine studies annually. Four out of five diagnostic-imaging procedures in nuclear medicine use this isotope. The parent molybdenum-99 (half-life = 66 hours) decays into the daughter nuclide technetium-99m (half-life = 6 hours).

Molybdenum-99 is recovered from uranium fission products after neutron irradiation in a high-flux reactor. Isotope targets for molybdenum-99 usually require further enrichment of uranium to make highly enriched (weapons-grade) uranium. Both handling and storage of subcritical amounts of uranium have to be carefully controlled. Neutron-irradiation research reactors are usually located at government laboratories and operate around the clock to produce molybdenum-99.



Brookhaven National Laboratory's first (circa 1950s) experimental technetium-99m generator with molybdenum-99 sorbed on alumina.

Most commercial molybdenum-99/technetium-99m generators use column chromatography, in which molybdenum-99 is sorbed onto alumina or alumina/silica phases. Contacting the solid phases with saline elutes the soluble technetium-99m, which results in a technetium-99m-containing saline solution that is then added in an appropriate concentration to the kits to be used.

Ideally, the eluting technetium-99m sample should be free of the long-lived parent to minimize the radiation dose to the patient. In reality, molybdenum-99 breakthrough occurs and has to be closely monitored. The Nuclear Regulatory Commission limits the breakthrough for molybdenum-99 contamination to 1 microcurie (μCi) of molybdenum-99 per millicurie (mCi) of technetium-99m, not to exceed 5 microcuries molybdenum-99 per dose. The technetium-99m elution efficiency varies between 75 and 90 percent of equilibrium activity, depending on the sorption system used.

The shelf life of a molybdenum-99/technetium-99m generator is about three half-lives, or approximately one week. Clinical nuclear-medicine units must purchase at least one such generator per week or order several in a staggered fashion. Researchers at Brookhaven National Laboratory developed the first experimental molybdenum-99/technetium-99m generator in the late 1950s.

Recently, concern has been raised about the future availability of the reactor-produced molybdenum-99 parent. Supply vulnerability of this isotope is primarily caused by two conditions. Just five research reactors produce about 95 percent of the worldwide demand for molybdenum-99. They are the Petten High Flux Reactor in the Netherlands; BR2 at Mol, Belgium; Osiris at Saclay, France; NRU at Chalk River, Canada; and the Safari-1 at Pelindaba, South Africa. These facilities are all more than four decades old. Several incidents in 2007 and 2008 and unexpected simultaneous maintenance outages of the aging research reactors caused critical shortages of molybdenum-99. On the other hand, the use of highly enriched uranium, the commonly used target material in the production of molybdenum-99, has come under increasing scrutiny because of proliferation concerns.

In view of these difficulties, alternative molybdenum-99 production methods are being sought. The NNSA has proposed studying three promising technologies: accelerators, solution reactors, and low-enriched uranium targets.

Strontium-82/rubidium-82 radionuclide generators

Positron emission tomography (PET) is superior to SPECT in the sense that it produces fewer artifacts, has a higher spatial resolution, and has the capability to perform quantitative measurements at the peak of stress and speed. PET scan times of 30 to 40 minutes mean more patients can be processed in a day, compared to the two or more hours it takes for a SPECT scan. The major advantage of SPECT, however, is that it's much more available and widely used and much cheaper than PET.

Strontium-82 (half-life = 25.5 days) is the parent of the CardioGen-82 PET Perfusion Imaging isotope rubidium-82 (half-life = 1.3 minutes), which has

been used in myocardial perfusion studies. The short half-life of rubidium-82 allows the sequential performance of scans every 10 minutes while minimizing the radiation dose to the patient. Available evidence suggests that myocardial perfusion PET provides an accurate means for diagnosing obstructive coronary arterial disease and appears superior to SPECT, especially in obese patients and in patients undergoing pharmacologic stress.

Germanium-68/gallium-68 radionuclide generators

Germanium-68 (half-life = 270.8 days) decays into gallium-68, a positron emitter (half-life = 67.6 minutes), which is used extensively in PET. Although some radionuclide generator principles have been suggested, there is no germanium-68/gallium-68 generator system approved by the U.S. Food and Drug Administration available on the market as yet.

In Europe, Eckert & Ziegler, for example, supplies a gallium-68 generator system on the basis of titanium(IV) oxide (the IGG100). A freshly supplied generator in full equilibrium typically yields approximately 70 percent gallium-68 for each elution, while the reported parent breakthrough for this system is 0.000007% per milliliters (mL) of elution volume.

At Los Alamos, a germanium-68/gallium generator system based on germanium sorption on organic exchanger resins has been under investigation. The major advantage in the use of organic resins is the minimization of trace metal leakage from the solid phase. Trace metals released into the eluting gallium-68 batch may interfere with subsequent radiolabeling.

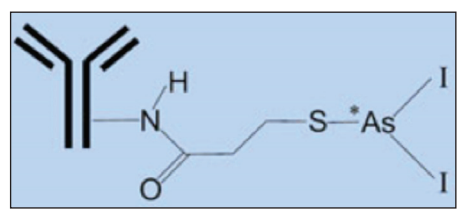
Selenium-72/arsenic-72 radionuclide generators

Selenium-72 (half-life = 8.5 days) is the parent of the short-lived positron emitter arsenic-72 (half-life = 26 hours). Arsenic radioisotopes, and arsenic-72 with its convenient half-life in particular, have strong imaging potential. The fact that this notorious element—especially in its trivalent state—strongly binds to sulfhydryl terminals of proteins is the underlying reason for its high toxicity. Ironically, this is the exact reason why radioarsenic, covalently bonded to an antibody, yields high-resolution images of tumors.

The development of a convenient selenium-72/arsenic-72 generator system suitable to a clinical setting has been hampered by various obstacles. To begin with, selenium-72 production and recovery is a complex process. Moreover, the close chemical behavior of parent (selenium) and daughter (arsenic) elements makes it difficult to find an elegant milking mechanism. The matter is further complicated by the occurrence of at least two species per element with different oxidation states, which poses a significant challenge to the labeling chemist.

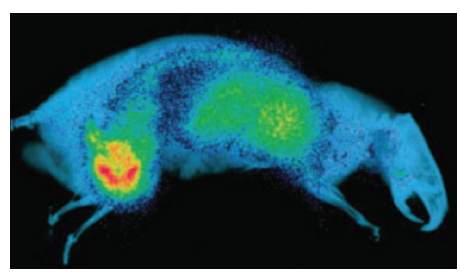
Dennis Phillips, formerly of Los Alamos, introduced and patented a distillation-type generator system in the 1990s, where the arsenic-72 activity is periodically distilled off the selenium-72 parent matrix. This system, however, requires a more elaborate radiochemical facility and trained personnel for operation. Other systems have been suggested; however, they are either tailored to very small

A CardioGen-82 generator unit.



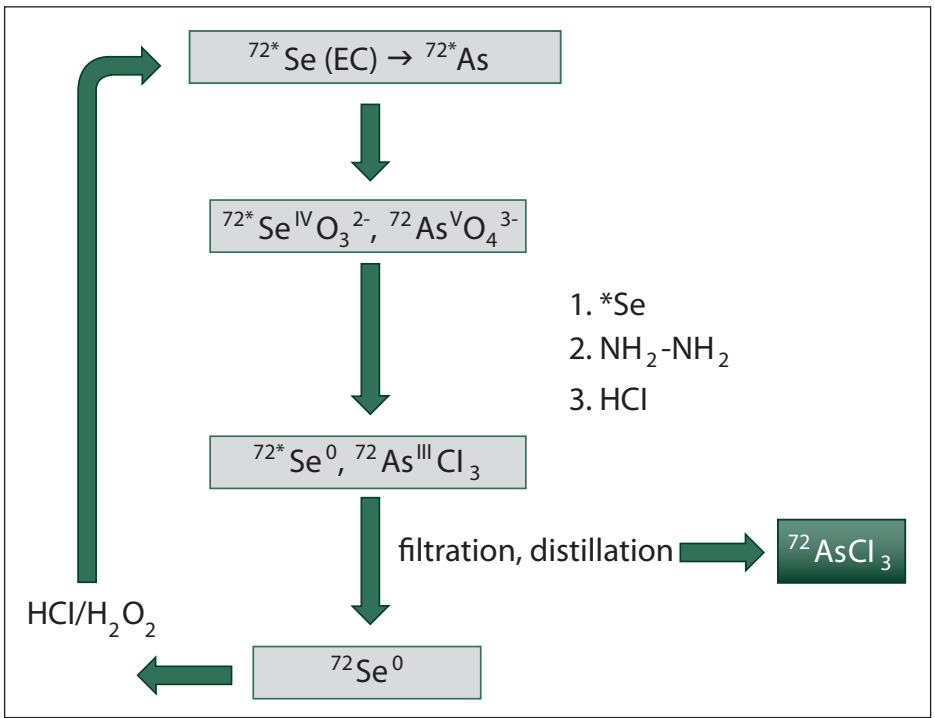
The bavituximab antibody labeled with arsenic.

Marc Jennewein et al., Clinical Cancer Research, March 2008



A positron emission tomography scan of a rat with tumors imaged with the new radioarsenic technique. An X-ray image of the rat is overlaid on the scan.

Marc Jennewein et al., Clinical Cancer Research, March 2008



Selenium-72/arsenic-72 generator based on distillation.

Further reading

"Manufacture of strontium-82/rubidium-82 generators and quality control of rubidium-82 chloride for myocardial perfusion imaging in patients using positron emission tomography," Teresa M. Alvarez-Diez et al., *Applied Radiation and Isotopes* 50, 1999.

"^{99}Mo supply—the times they are a-changing," Dewi M. Lewis, *European Journal of Nuclear Medicine and Molecular Imaging* 36, 2009.

"Production of strontium-82 for the Cardiogen® PET generator: a project of the Department of Energy Virtual Isotope Center," Dennis R. Phillips et al., *Radiochimica Acta* 88(3-4), 2000.

(tracer) quantities of loading activity or require a set of hazardous chemicals for the milking step.

Efforts have been launched by Los Alamos scientists in collaboration with University of Missouri faculty to develop a selenium/arsenic separation based on selective chelation. Selective chelators, attached to a polymer backbone, could achieve an efficient separation of ingrown arsenic activity from its selenium parent under relatively mild conditions without the introduction of problematic reagents.

Conclusion

Radionuclide generators have become an indispensable part of state-of-the-art medical imaging capabilities. They enable the clinician to use short-lived radioisotopes without needing a close-by radionuclide production facility. Although there are many parent–daughter pairs that could potentially find use in clinical purposes, only a few generator systems have achieved wider acceptance so far. Technetium-99m is still the most widely used medical nuclide, and—once supply shortages are overcome—it will most likely remain the most popular tool in the world of nuclear imaging owing to both its half-life and chemical separation simplicity.

For PET, on the other hand, several generator systems may become important in the future. While the strontium-82/rubidium-82 system has already been routinely used, the germanium-68/gallium-68 system receives increasing attention. The availability of longer-lived positron emitters may shift the focus of imaging from rapid-uptake mechanism to the visualization of longer-term metabolic processes. Short-lived arsenic-72 could become an important tool to reach this goal once a marketable system has been developed.

Recovery and Uses of Americium-241

This article was contributed by Louis Schulte of the Actinide Process Chemistry Group, Toby Vigil of Program Management, David Pesiri of the Technology Transfer Division, and Kevin John of the Office of Science Programs.

It's now some 65 years since the discovery of americium by Glenn Seaborg and his team at the wartime Metallurgical Laboratory in Chicago (later to become Argonne National Laboratory). The discovery of elements number 95 and 96 (americium and curium, respectively) was somewhat famously first publicly disclosed by Seaborg on a radio program called "Quiz Kids" in November 1945.

The initial americium isotope discovered was americium-241 (half-life = 432.7 years), which along with the longest-lived isotope, americium-243 (half-life = 7380 years), comprise the most important and useful of the americium isotopes. These two isotopes (along with smaller amounts of americium-242 and americium-242m; m = metastable) are found together in irradiated reactor fuel in quantities and ratios dependent on burnup in the nuclear reactor.

High isotopic purity americium-241 can be recovered from plutonium that has been separated from the irradiated fuel and then allowed to "age" before separation of ingrowth americium-241. Both weapons-grade plutonium and reactor-grade plutonium isotopic mixtures include a fraction of plutonium-241, with reactor-grade having a greater fraction of the heavier plutonium isotopes. The beta decay of plutonium-241 (half-life = 14.4 years) to americium-241 provides a path to high isotopic purity material in separated plutonium.

Glenn Seaborg (far left) and Quiz Kids Sheila Conlon and Bob Burke (aka actor Robert Easton) when Seaborg informally announced the discovery of element 95 (americium) and element 96 (curium) on the radio quiz show.

Cowan, sponsor of "Quiz Kids"

Uses of americium-241

Americium-241 has been considered by Seaborg and others to be among the most useful of the actinide elements. The reasons are availability, a useful half-life, and nuclear decay by nearly monoenergetic alpha emissions—5.44 and 5.49 megaelectronvolts (MeV)—with an associated high yield (30 percent) of 59.6-kiloelectronvolt (keV) gamma emissions.

A few higher-energy gamma emissions are observed for americium-241 but are small in quantity compared to the predominant 59.6 keV signature. This relatively low-energy gamma emission finds use in a wide variety of applications including gamma transmission/absorption and backscatter measurements (primarily for determination of thickness of materials/coatings, density and radiographic measurements). The alpha emission finds use in measurement of thin films, gas densities, and as a source of alpha ionization. Both the alpha and gamma signatures find use in calibration of instrumentation.

The most common and practical use of americium-241 is in the millions of smoke detectors in use throughout the world (see sidebar on page 23). An additional ongoing need for larger quantities of americium-241 is for use in americium-beryllium (AmBe) neutron sources. The value of AmBe neutron sources is that they provide a portable, tough, continuous, and reliable source of neutrons outside a reactor or a neutron generator. Mixtures of finely divided americium-241 oxide and beryllium metal powder create a neutron source from "alpha-N" reactions (shorthand representation $^{9}Be(\alpha,N)^{12}C$).

Acknowledgements

The authors would like to thank the following people and organizations for their support:

Wolfgang Runde, manager, DOE National Isotope Program (SPO-SC)

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Actinide Process Chemistry (PMT-2), Kent Abney, group leader

Jim Rubin (PMT-2) and Bill McKerley (PM-2)

PMT-2 Aqueous Chloride team:

Cindy Kowalczyk, Jennifer Butler, Steven Archuleta, Kennard Wilson, and Ronnie Chavez are working to complete hardware modifications to CLEAR gloveboxes.

Steve McKee devised the original synthesis of di-(4-t-butylphenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide. This synthesis was later scaled larger by Dick Bartsch (Texas Tech) and Keith Pannell (UTEP) and coworkers.

Richard Salazar performed demonstration runs of actinide removal.

Recent efforts to update the P&ID and glovebox installation have been led by Jay Rutten, with support from Chris Thorn and Sheldon Apgar.

Many others have provided support, including Raj Vaidya, Joe Jackson, Simon Balkey, Drew Kornreich, Mark Robinson, Wayne Smyth, Mark Dinehart, Vance Hatler, Jacque Seay, Cecil Brown, Keith Fife, Marlayne Mahar, and Anthony Lupinetti.

Americium-241 has a half-life (432 years) useful for sources because this relatively high-specific activity (3.42 curies per gram) provides good neutron-source efficiency, useful neutron flux ranges, and steady neutron output for years in an AmBe source. Beryllium has the highest neutron yield from (α,N) reaction of any element, although even beryllium requires many thousands of alpha particles to produce one neutron.

The largest AmBe neutron sources, and the majority of these sources, are used for oil and natural gas well-logging purposes. Approximately 4000 oil and gas drill rigs exist throughout the world, and worldwide exploration is a multi-billion-dollar annual industry. Logging wells (density and porosity among other characteristics) is an important step to ascertain the predicted production characteristics. Well logging with a neutron source may be "wire line" in an open or cased hole, or "Logging-While-Drilling" where the neutron source and detector(s) are on board a special section of the drilling stem (see sidebar on page 24). AmBe is also used in thickness-gauging measurements of light alloys, glass, plastics, and rubber; and in nondestructive elemental analysis applications.

Separations of americium-241

Following separations work on microgram-scale at the Metallurgical Laboratory in Chicago in the late 1940s, gram-scale americium-241 separations work and many chemical speciation studies were done at Los Alamos in the early 1950s. The first external sales of americium-241 recovered at Rocky Flats occurred in 1962 through the Oak Ridge National Laboratory National Isotope Center, a business office run for the DOE Isotope Program. Larger-scale americium-241 separations efforts (about kilogram annual quantities) occurred at the Rocky Flats and Hanford sites throughout the 1960s and 1970s as part of plutonium reprocessing campaigns of both weapons-grade and reactor-grade plutonium.

From 1978 to 1984, larger quantities of americium-241 were recovered as part of plutonium reprocessing campaigns at Los Alamos and the Savannah River Site (SRS). These campaigns used aged, reactor-grade plutonium as feedstock, and produced plutonium oxide (PuO_2) for the Fast Flux Test Facility in Hanford as well as other DOE programs. Los Alamos efforts produced about 6.5 kg of americium-241, while SRS produced about 8 kg of americium-241.

The large inventory of americium-241 stockpiled from these campaigns and earlier production resulted in a surplus of material, with little interest in additional recovery of material in the United States for many years. The issues of handling americium-241 are part of normal plutonium reprocessing operations, and without a demand, americium-241 has gradually become considered a troublesome byproduct of plutonium reprocessing rather than a useful resource. The pool of available americium-241 isotope in the United States has been depleted to zero over the intervening years, and there is no current domestic supplier for new material.

The americium-241 ingrowth content in plutonium varies with the time period since plutonium was last separated and with the atomic fraction of

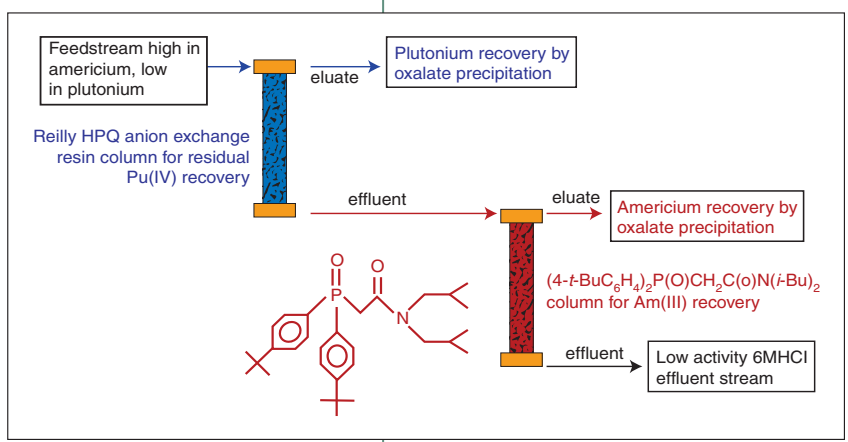
plutonium-241, which is significantly higher in reactor-grade material. As two or more half-lives of plutonium-241 have gone by since the weapons plutonium originally came out of the reactor (most of this plutonium was made in the 1950s, 1960s, and 1970s), ingrowth of future americium-241 after purification today will inherently be much smaller.

Proposed future americium-241 recovery work

Ingrowth of americium-241 in separated plutonium provides a path to high isotopic purity material, and as a result the opportunity for recovery of new americium-241 is closely tied to plutonium reprocessing operations. In the United States, reprocessing of plutonium already separated from irradiated fuel occurs only within the DOE weapons complex, with Los Alamos having the major role as the Plutonium Center of Excellence.

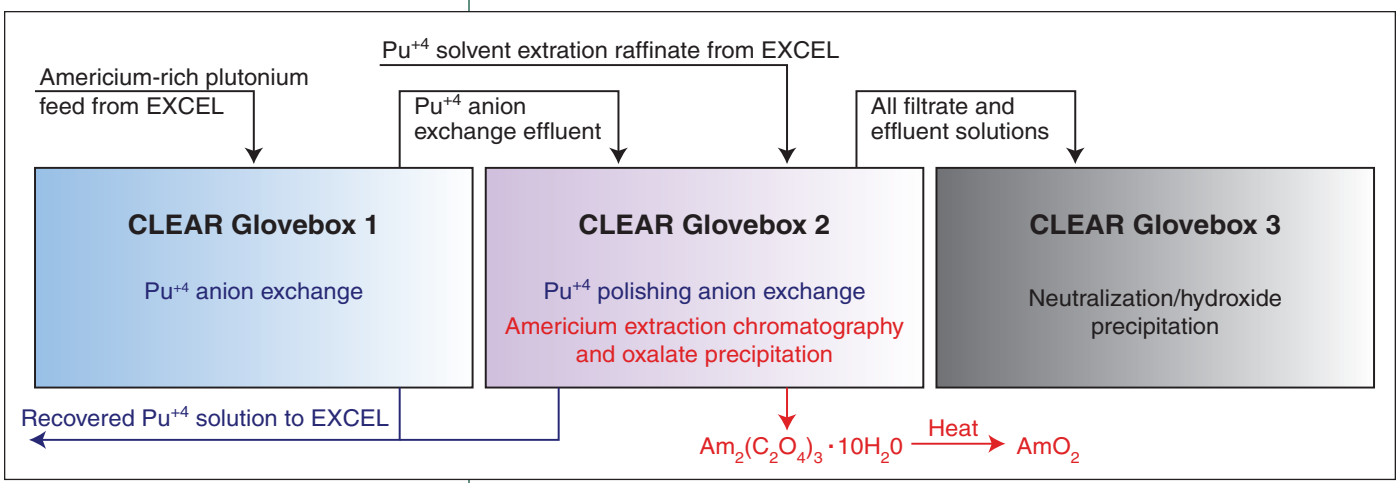
Los Alamos does reprocessing of plutonium to support a limited pit-manufacturing capability, vault consolidation, and has produced high-purity PuO_2 for mixed oxide fuel (MOX) and other programs. Existing operations in plutonium pyrochemistry reprocessing make use of a step early in the flowsheet for americium extraction (also called molten salt extraction) to reduce gamma dose to operators in subsequent processing steps.

The partition in the americium extraction step concentrates the americium about 20-fold in a small amount of plutonium, creating residue items rich in americium that could be used as an efficient feedstock for a campaign to recover high-purity americium-241. The large number of these americium-241-enriched residues in current vault holdings, along with ongoing future plutonium reprocessing work, would allow high-purity americium-241 recovery at a large scale (500–750 grams per year) for several years.



Improvements to plutonium reprocessing efforts at Los Alamos have been focused on operational efficiency and on minimizing the volume and hazards associated with solid wastes and liquid effluents. Additional goals have been to reduce dose to glovebox operators and waste handlers, with dose issues often tied to gamma exposure from the 59.6-keV emission of americium-241. Technology was developed to reduce losses of actinides in aqueous process effluent streams, which provided benefit in reduced hazard and volume of actinide residues stored in a protected vault, and in the quantity of actinides discarded to the Waste Isolation Pilot Plant (WIPP) in southern New Mexico.

The Chloride Extraction and Actinide Recovery (CLEAR) glovebox line was designed primarily for waste volume and hazard minimization, operator dose reduction, and improved process efficiency in support of existing hydrochloric



Further reading

"Americium, Its Early History and Gram-Scale Separation," Robert A. Penneman, *Americium and Curium Chemistry and Technology, Topics in f-Element Chemistry,* 1985.

"The 40th Anniversary of the Discovery of Americium and Curium," Glenn T. Seaborg, *Americium and Curium Chemistry and Technology, Topics in f-Element Chemistry,* 1985.

"The Most Useful Isotope of the Actinide Elements: Americium-241," James D. Navratil, Wallace W. Schulz, and Glenn T. Seaborg, *Journal of Chemical Education*, 1986-1990.

"Chapter 8: Americium," Wolfgang H. Runde and Wallace W. Schulz, *The Chemistry of the Actinide and Transactinide Elements, Third Edition*, 2006.

"The First Isolation of Americium in the Form of Pure Compounds; Microgram-Scale Observations on the Chemistry of Americium," B.B. Cunningham, *The Transuranium Elements: Research Papers,* 1949.

acid (HCl) process operations. Because of changes in mission needs and priorities, the process was never fully implemented. The demonstrated process used an extraction chromatography resin loaded with $(4\text{-}t\text{-}BuC_6H_4)_2P(O)CH_2C(O)N(i\text{-}Bu)_2$ [di-(4-t-butylphenyl)-N,N-diisobutylcarbamoylmethyl-phosphine oxide] as a ligand for trivalent actinides.

Recovery of purified americium-241 oxide to meet specification requirements was not an important driver in original demonstration runs, but data are supportive of this capability. Modest alteration of the demonstrated technology would allow recovery of pure americium-241 from pyrochemistry residues processed in existing hydrochloric acid plutonium recovery operations. This combination of available feedstream, technology, and available infrastructure makes the CLEAR glovebox line a viable choice to support domestic production of high-purity americium-241 oxide.

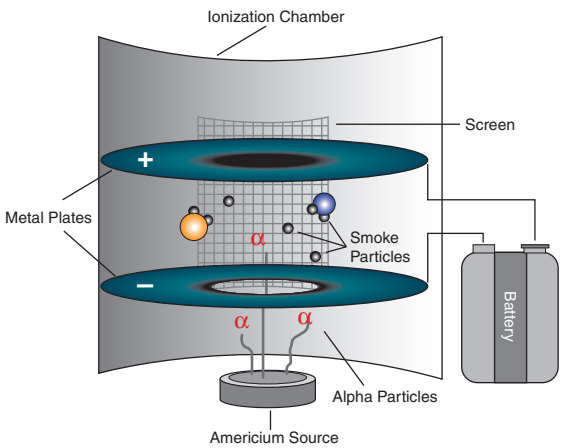
Am-USA

Los Alamos is in the process of building a capability to separate clean americium-241 from existing residues and plutonium reprocessing operations for sales and distribution by the DOE National Isotope Program. Am-USA is the vehicle through which Los Alamos will connect these internal benefits with the commercial value of americium-241 in the marketplace. Am-USA is led by the authors of this article, who represent the TA-55 Program Office, TA-55 Plutonium Manufacturing Technology, Los Alamos Technology Transfer, and the Los Alamos Isotope Program; and coordinated with the DOE National Isotope Program.

The goals of this project are to develop the capability at Los Alamos to produce commercial quantities of americium-241, to promote the safe and responsible use of americium-241, and establish a secure and reliable source of americium-241 for the benefit of U.S. industry.

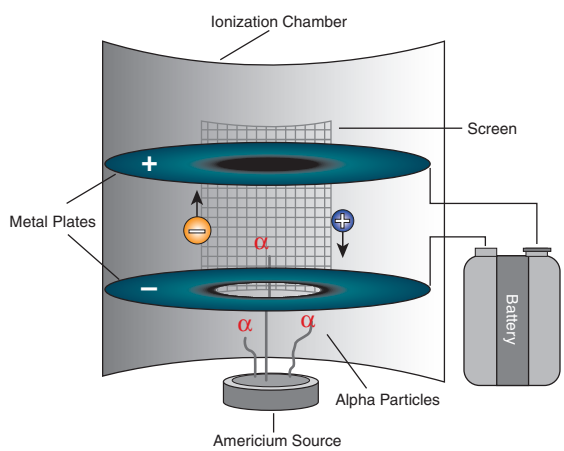
Ionization technology

Alpha particles from the americium source ionize air molecules.



In the smoke-free chamber, positive and negative ions create a small current as they migrate to charged plates.

Smoke particles and combustion gases interact with the ions generated by the alpha particles, restoring them to their neutral electronic state and decreasing the electrical current passing through the cell.



As fewer ions are available to migrate to the plates, the disrupted current triggers the alarm.

Environmental Protection Agency, www.epa.gov

How Smoke Detectors Work

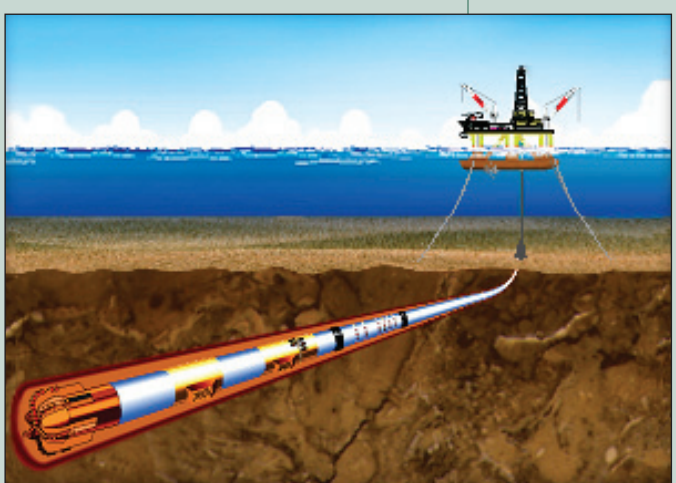
Ionization sensor smoke alarms contain a small amount of radioactive material, americium embedded in a gold foil matrix within an ionization chamber. The matrix is made by rolling gold and americium oxide ingots together to form a foil approximately one micrometer thick. This thin gold–americium foil is then sandwiched between a thicker (about 0.25 millimeter) silver backing and a 2-micron-thick palladium laminate. This is thick enough to completely retain the radioactive material but thin enough to allow the alpha particles to pass.

The ionization chamber is basically two metal plates a small distance apart. One of the plates carries a positive charge, the other a negative charge. Between the two plates, air molecules—made up mostly of oxygen and nitrogen atoms—are ionized when electrons are kicked out of the molecules by alpha particles from the radioactive material (alpha particles are big and heavy compared to electrons). The result is oxygen and nitrogen atoms that are positively charged because they are short one electron; the free electrons are negatively charged.

The diagrams illustrate how ionization technology works. The positive atoms flow toward the negative plate, as the negative electrons flow toward the positive plate. The movement of the electrons registers as a small but steady flow of current. When smoke enters the ionization chamber, the current is disrupted as the smoke particles attach to the charged ions and restore them to a neutral electrical state. This reduces the flow of electricity between the two plates in the ionization chamber. When the electric current drops below a certain threshold, the alarm is triggered.

Well Logging

Logging measurements are quite sophisticated. The prime target is the measurement of various geophysical properties of the subsurface rock formations. Of particular interest are porosity, permeability, and fluid content. Porosity is the proportion of fluid-filled space found within the rock. It is this space that contains the oil and gas. Permeability is the ability of fluids to flow through the rock. The higher the porosity, the higher the possible oil and gas content of a rock reservoir. The higher the permeability, the easier for the oil and gas to flow toward the wellbore. Logging tools provide measurements that allow for the mathematical interpretation of these quantities.



Large americium-beryllium neutron sources are used for oil and natural gas well logging. In a logging-while-drilling application, the neutron source and detector are on board a section of the drilling stem.

AccuTech: The MWD Company

Neutrons are typically emitted by a chemical source such as americium-beryllium (AmBe) or plutonium-beryllium (PuBe). Fast neutrons are emitted by these sources with energy ranges from 4–10 megaelectronvolts (MeV), and inelastically interact with matter. Once slowed down to 2 MeV, they start to scatter elastically and slow down further until the neutrons reach a thermal energy level of about 0.025 eV. When thermal neutrons are then absorbed, gamma rays are emitted. A suitable detector, positioned at a certain distance from the source, can measure either epithermal neutron population, thermal neutron population, or the gamma rays emitted after the absorption.

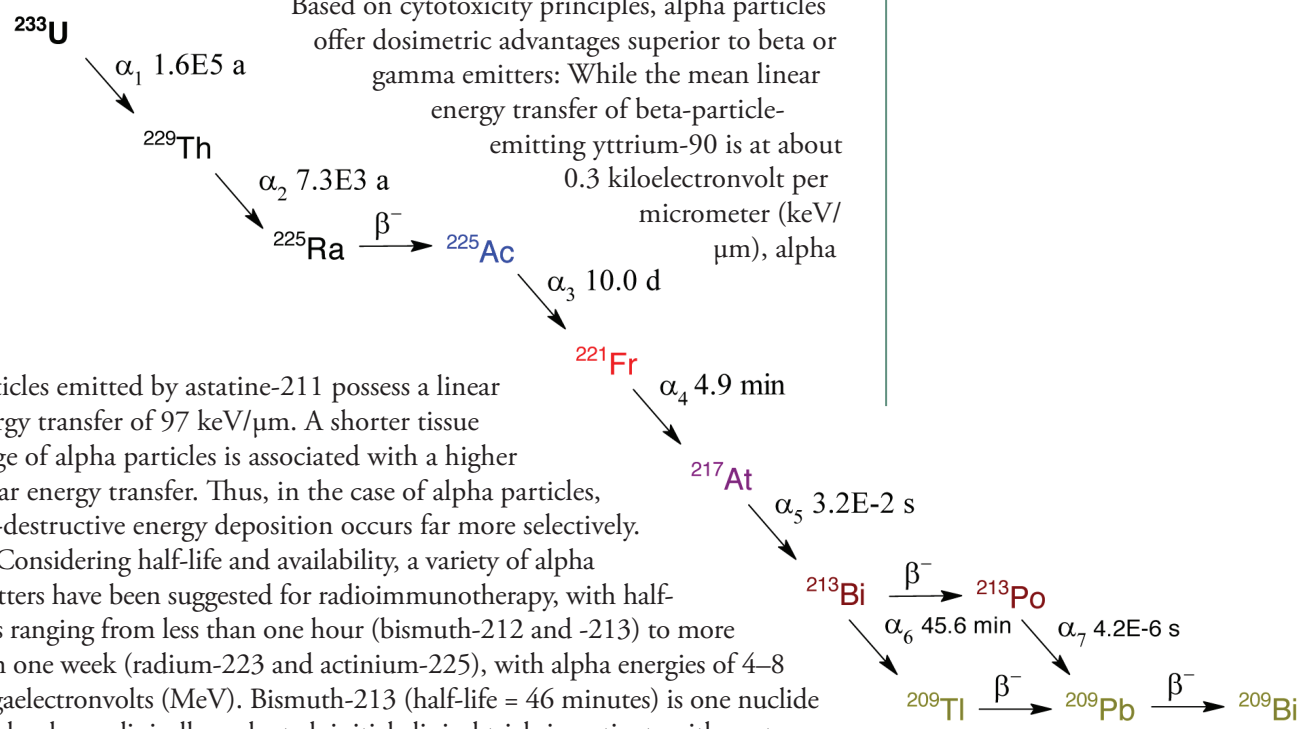
Mechanics of elastic collisions predict that the maximum energy transfer occurs during collisions of two particles of equal mass. Therefore, a hydrogen atom will cause a neutron to slow down the most as they are of roughly equal mass. As hydrogen is fundamentally associated with the amount of water and/or oil present in the pore space, measurement of neutron population within the investigated volume is directly linked to porosity.

Actinium-225: A Promising Alpha Emitter for Particle-Mediated Radioimmunotherapy

This article was contributed by Michael Fassbender of the Inorganic, Isotope and Actinide Chemistry Group.

Radioimmunotherapy makes use of the cytotoxicity delivered by particles with a high linear energy transfer. Alpha particles are monoenergetic helium nuclei (He-4) with a rather high kinetic energy, which are emitted during the radioactive decay of certain heavy-element nuclei. There are approximately 100 unstable, radioactive nuclei that decay with the emission of He-4 particles. Predominately among those alpha emitters are elements with atomic numbers greater than or equal to 82 (lead and elements heavier than lead).

Based on cytotoxicity principles, alpha particles offer dosimetric advantages superior to beta or gamma emitters: While the mean linear energy transfer of beta-particle-emitting yttrium-90 is at about 0.3 kiloelectronvolt per micrometer (keV/µm), alpha particles emitted by astatine-211 possess a linear energy transfer of 97 keV/µm. A shorter tissue range of alpha particles is associated with a higher linear energy transfer. Thus, in the case of alpha particles, cell-destructive energy deposition occurs far more selectively.



Thorium-229 decay chain leading to (stable) bismuth-209 via actinium-225 and bismuth-213.

Considering half-life and availability, a variety of alpha emitters have been suggested for radioimmunotherapy, with half-lives ranging from less than one hour (bismuth-212 and -213) to more than one week (radium-223 and actinium-225), with alpha energies of 4–8 megaelectronvolts (MeV). Bismuth-213 (half-life = 46 minutes) is one nuclide that has been clinically evaluated; initial clinical trials in patients with acute myeloid leukemia have demonstrated the effectiveness of bismuth-213 in killing cancer cells. Bismuth-213 emits 8-MeV alpha particles with a tissue penetration of about 100 µm, corresponding to six to ten nearby cell layers.

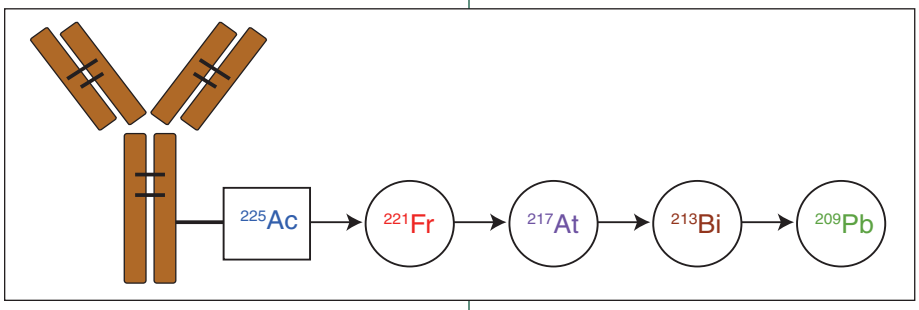
However, there are significant limitations associated with this isotope due to its short half-life. A favorable half-life should allow for radionuclide production, radiolabeling of the target molecule, administering a dose, and sufficient time for in vivo distribution. The complete process takes far more than one bismuth-213 half-life, thus decreasing the therapeutic dose administered to the patient.

Bismuth-213 is a generator-produced nuclide; actinium-225 (half-life = 10 days) is its parent. Actinium-225 itself presents an attractive alternative to its short-lived daughter, bismuth-213. It is a member of the thorium-229 decay scheme and emits four alpha particles during the decay cascade until a pure beta emitter, thallium-209 (half-life = 2.2 minutes), is produced. Thallium-209, in turn, decays to lead-209 (half-life = 3.3 hours) and finally to bismuth-209, which is stable. There are six daughters of actinium-225, which are produced in

the cascade to stable bismuth-209. Each actinium-225 decay is succeeded by five alpha and three beta disintegrations, most of high energy.

Actinium-225 cytotoxicity has been compared to bismuth-213 in vitro using identical target molecules either labeled with bismuth-213 or actinium-225, revealing that much less actinium-225 radioactivity is required to reach a median lethal dose, most probably owing to longer half-life and multiple alpha emissions. The enhanced potency of actinium-225 as compared to bismuth-213 has also been demonstrated in vivo in a model of prostate cancer. The suitability of actinium-225 as an internal radiation source, however, is closely tied to the availability of strong chelators, which can be functionalized such as to bind to biomolecules.

To harness decay daughter radiation, chelation systems should not only be capable of strongly binding the parent actinium in the biological target tissue, but also its alpha-decay daughters. One system to accommodate these requirements—the "nanogenerator"—was suggested by Michael McDevitt and colleagues in a paper titled "Tumor therapy with targeted atomic nanogenerators," which was published in *Science* in 2001.



The actinium-225 "nanogenerator" system. The alpha particle recoil energy is likely to thrust the decay products out of the chelate.

The nanogenerator focuses on strongly coordinated actinium-225 for target-cell delivery, internalizing the actinium-225 bioconjugate into the target cell, and retaining decay daughters within the target cell. One has to bear in mind that actinium-225 decay daughters are unlikely to remain bound to the initial chelator because the alpha-particle recoil energy, which is usually between 100 and 200 keV, will exceed the binding energy of a macrocyclic metal-ion complex.

Actinium-225 production and supply

Actinium-225 can be obtained either from the decay of uranium-223 or from successive neutron-capture reactions/beta-decay combinations starting with radium-226, which eventually yields thorium-229.

^{226}Ra (n,γ) ^{227}Ra (β^-, 42.2 minutes) → ^{227}Ac (n,γ) ^{228} Ac (β^-, 6.1 hours) → ^{228} Th (n,γ) ^{229}Th

The yield of thorium-229 via radium-226 (n,γ) is about 150 millicuries per gram (mCi/g) of radium-226 for twenty-four days at a neutron flux of a quadrillion (10^{15}) neutrons per square centimeter per second (cm^2s) with a thermal to epithermal ratio of 10. Actinium-225 can also be produced via a charged-particle-induced, low-energy reaction on radium-226. Another path leading to thorium-229 and actinium-225 is the use of high-energy protons on natural thorium-232, which is currently under investigation at Los Alamos. The paths, or nuclear reaction channels, are shown in a green box on page 27.

Actinium-225 that has been used in Phase I and II clinical trials is presently produced routinely at Oak Ridge National Laboratory and at the Institute for

Transuranium Elements in Karlsruhe, Germany. At Oak Ridge, actinium-225 is produced via the decay of thorium-229 (half-life = 7340 years). The two daughter isotopes, radium-225 (half-life = 14.8 days) and actinium-225 (half-life = 10 days), are initially separated from the parent via anion exchange. Larger batches of actinium-225 are freshly separated from thorium-229, while a stock of purified radium-225 provides an additional continuous supply for smaller batches of the radioisotope.

As an example, the Sloan Kettering Memorial Cancer Center, where actinium-225 is used as an in-house generator for bismuth-213, receives up to 50 millicuries of actinium-225 originating from direct separation, while 20 millicuries or less are recovered from purified radium-225.

Thorium-229, in turn, originates from uranium-233, which was produced via neutron capture using a natural thorium-232 target as part of the U.S. molten salt breeder reactor research program.

$^{232}Th\ (n,\gamma)\ ^{233}Th\ (\beta^-,\ 22.3\ minutes) \rightarrow\ ^{233}Pa\ (\beta^-,\ 27\ days) \rightarrow\ ^{233}U$

Oak Ridge and the Institute for Transuranium Elements use uranium-233 as a source for thorium-229. Uranium-233 is currently stored at Oak Ridge. Considering the theoretical specific activity of thorium-229 (0.2 curies per gram), the entire Oak Ridge uranium-233 stockpile can only yield about 8 curies of extracted thorium-229, which will not be enough to meet the anticipated actinium-225 and bismuth-213 market demand in the near future. Oak Ridge's thorium-229 supply is about 150 millicuries.

Besides the limited uranium-233 stockpile, routine recovery of thorium-229 from fissile uranium-233 does not seem feasible, taking into account both the low in-growth rate and increasing nuclear safeguards regulations. At present, uranium-233 is the only viable source for thorium-229 and its decay progenies.

Actinium-225 chemistry at Los Alamos

The production and chemistry of actinium-225 has been under investigation at Los Alamos both from the standpoint of its alternative production using a proton beam at the Los Alamos Isotope Production Facility (IPF) and of effectively binding it to biomolecules.

High-energy proton bombardment of thorium-232 results in at least five different nuclear reaction channels with desirable residual nuclei.

$^{232}Th(p,4n)\ ^{229}Pa\ (\beta^+,\ 1.4\ d) \rightarrow\ ^{229}Th\ (\alpha,\ 7340\ a) \rightarrow\ ^{225}Ra\ (\beta^-,\ 14.8\ d) \rightarrow\ ^{225}Ac$

$^{232}Th(p,p3n)\ ^{229}Th\ (\alpha,\ 7340\ a) \rightarrow\ ^{225}Ra\ (\beta^-,\ 14.8\ d) \rightarrow\ ^{225}Ac$

$^{232}Th(p,4n)\ ^{229}Pa\ (\alpha,\ 1.4\ d) \rightarrow\ ^{225}Ac$

$^{232}Th\ (p,x)\ ^{225}Ac$

$^{232}Th(p,x)\ ^{225}Ra\ (\beta^-,\ 14.8\ d) \rightarrow\ ^{225}Ac$

One major byproduct is likely to be radium-224.

$^{232}Th(p,\alpha 5n)\ ^{224}Ac\ (EC,\ 2.9\ h) \rightarrow\ ^{224}Ra\ (\alpha,\ 3.7\ d)$

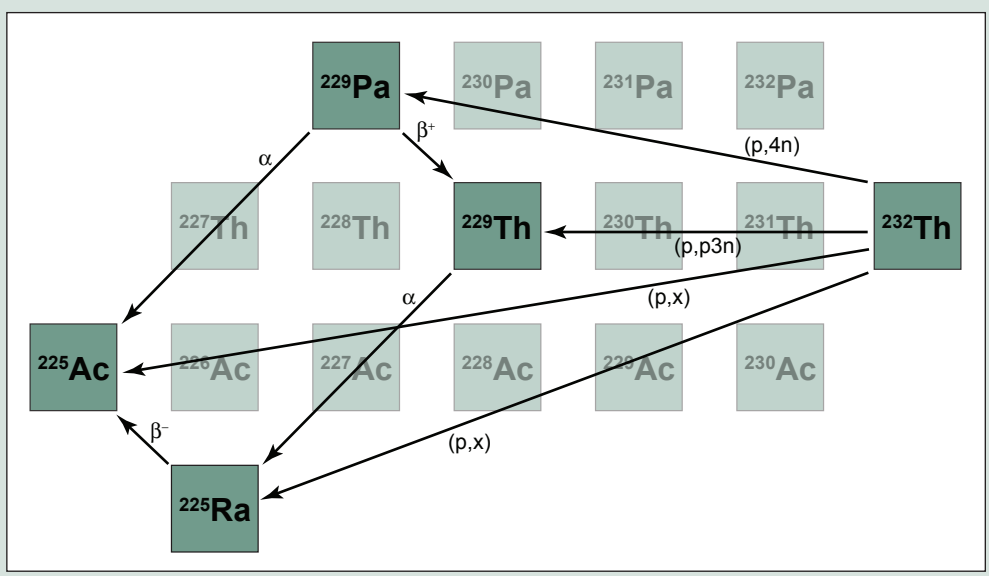
To establish an alternative production route for actinium-225, it is viable to compare production yields and contamination levels to the established

uranium-233 → thorium-229 → actinium-225 route. Production yields and contamination levels can only be accurately predicted upon the basis of nuclear reaction cross sections for each channel, and cross-section measurement is the primary objective at Los Alamos at this time.

Product recovery and purification will depend on a reliable radiochemical separation procedure for the four-element system of thorium, radium, actinium, and protactinium. This is not only the case for a later large-scale production program, but also to aid cross-section measurement.

Starting with a proton-irradiated thorium-232 target, the first separation step requires rapid chemical separation of protactinium-229 from the thorium target matrix before it further decays to thorium-229, to allow for separate cross-section measurement for the first and third nuclear reaction channels shown in a green box on page 27.

Nuclear reaction scheme



Nuclear scientists employ a special type of shorthand (used in equations shown in the green boxes on previous pages) to denote nuclear reactions where there is a combination of incident (or bombarding) and emitted particles to produce new isotopes. With the figure at right, we take the reader through the various equations in a more pedagogical fashion. Bombardment of thorium-232 (^{232}Th) with protons (the dark green box at the far right in the figure) produces four predominant reactions. One reaction is (p,4n) which denotes a reaction in which the thorium-232 nucleus absorbs one proton and emits four neutrons to give protactinium-229 (^{229}Pa). Once formed, the protactinium-229 can subsequently decay by either loss of an alpha particle (α) to give actinium-225 (^{225}Ac), or loss of a positron (β^+, electron capture) to give thorium-229 (^{229}Th). Proton bombardment of thorium-232 can also produce thorium-229 directly via a (p,p3n) reaction wherein the nucleus absorbs one proton and then emits one proton and three neutrons. The thorium-229 produced by either the (p,4n) or (p,p3n) reactions decays by loss of an alpha particle to produce radium-225 (^{225}Ra), which undergoes beta particle decay (β^-) to generate actinium-225. At higher incident proton energies, various combinations of protons, neutrons, deuterons, and alpha particles may be emitted after the absorption of the proton to form actinium-225 directly via (p,x)-type reactions. Finally, radium-225 may also be produced via a (p,x)-type reaction, after which it decays through the emission of a beta particle to form actinium-225.

This procedure must take no longer than one half-life (1.4 days), including target transportation from IPF to the chemical laboratory. Subsequent separation of actinium from radium is not so time critical due to longer half-lives. The proposed technique to separate lead traces from thorium mass is based on liquid–liquid extraction, where the thorium metal is dissolved in hydrochloric acid, converted to pyrosulfate, and extracted with diisobutylcarbinol.

Alternatively, anion and cation exchange could be used. "Off-line" decay of separated protactinium-229 to thorium-229, however, will not be a major contributor to thorium-229 formation owing to the very small ratio of the protactinium-229 half-life:thorium-229 half-life = 5.2×10^{-7}.

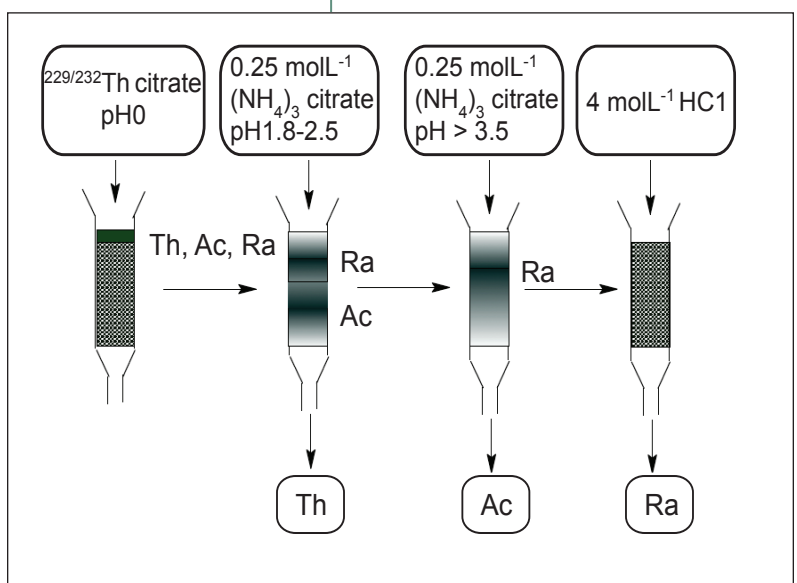
Nanogenerator application

Actinium-225 chelation research efforts at Los Alamos have been focusing both on coordinating actinium itself and retaining alpha-decay daughter isotopes. As mentioned before, successful actinium chelation does not necessarily retain both actinium-225 and the daughter products because of the high alpha-recoil energy. This leads to undesired daughter migration into non-target organs and associated radiotoxicity in healthy tissue. One approach, as pursued by Mary Barr of Los Alamos' Applied Engineering and Technology Division, is the introduction of chelating polymers with multiple coordination sites.

Another approach to limit daughter migration, is the use of nonchelating polymers in conjunction with the introduction of primary and secondary chelators. The primary chelator coordinates actinium-225, while secondary chelators are present to coordinate daughter nuclides of the decay chain. Jonathan Fitzsimmons and Robert Atcher discussed the approach in an article titled, "Synthesis and evaluation of a water-soluble polymer to reduce Ac-225 daughter migration," which was published in 2007 in the *Journal of Labelled Compounds and Radiopharmaceuticals*.

A combination of 1,4,7,10-tetraazacyclododecane-N, N′, N′, N′′-tetraacetic acid (DOTA, primary chelators to bind actinium-225), as well as secondary chelators acetate and diethylene triamine pentaacetic acid (DTPA), was used in conjunction with polyethylenimine (PEI) polymer derivatives. These highly branched polymer derivatives are rather resistant to radiolytic degradation. They serve as recoil-energy absorbers to allow reaction between daughter nuclides and secondary chelators.

Daughter retention as a function of time (range = 0–120 hours) was established for isotopes francium-221, bismuth-213, and thallium-209. Data indicate that the polymer derivatives sufficiently retained daughter



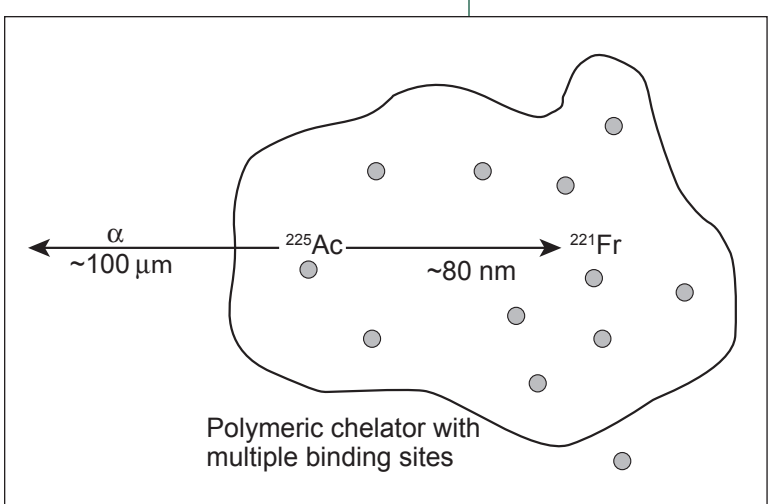
Possible cation exchange sequence for thorium/actinium/radium separation based on citrate chelation.

isotopes, rendering this post-labeling method capable of reducing daughter migration in vivo.

A synthetic procedure to make a bioconjugate based on this method would follow these steps:

- addition of a protected sulfhydryl to the PEI,
- addition of the primary chelator to the PEI,
- radiolabeling with actinium-225,
- addition of secondary chelators containing
- phosphate groups or carboxylate groups to the PEI,
- deprotection of the sulfhydryl, and
- formation of the thiol-maleimide linkage to the targeting molecule.

Further research will be conducted to determine daughter retention by PEI conjugates with targeting molecules.



Polymeric chelators may dissipate recoil energy and retain daughter nuclei.

Conclusion

Heavy-element alpha emitters show promise for the treatment of disseminated diseased tissue because they combine high cytotoxicity with an irradiation range of only a few cells. Several decay chains have multiple alpha emissions, thus offering the promise of increased cytotoxicity if the daughter nuclei can be retained in close proximity to the target cell. The radionuclide actinium-225 combines several favorable properties relevant to particle mediated radioimmunotherapy applications: favorable half-life of 10 days, high alpha-particle energy, versatile coordination chemistry, and several alpha-emitting daughter isotopes.

The availability of the isotope, however, is currently limited and largely dependent on stockpiled fissile nuclear material. Nuclear safeguards regulations and radiochemical aspects pose obstacles for the long-term future supply of actinium-225. Another hurdle to overcome is the effective retention of its decay daughters in the target tissue to avoid activity migration into and unintended damage to healthy organs.

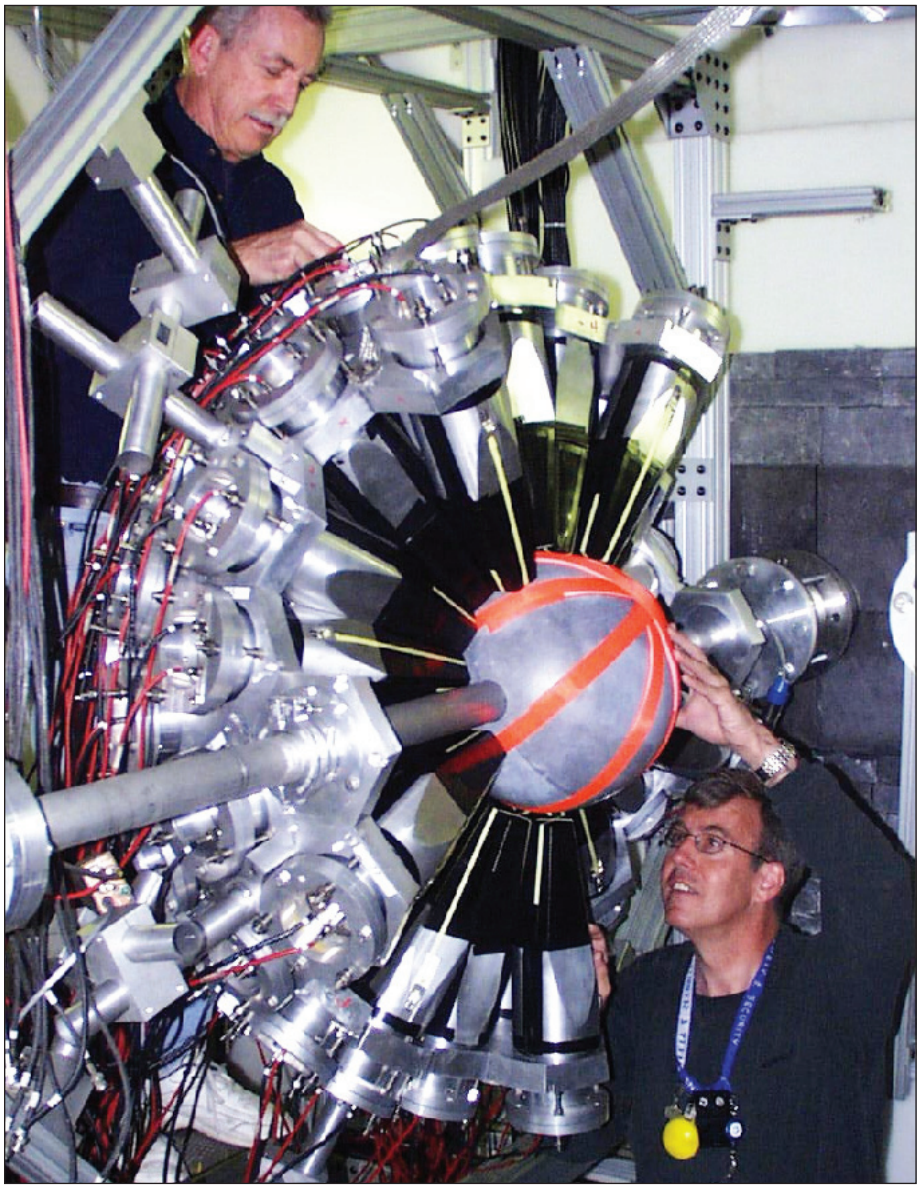
Current research and development at Los Alamos seek to strengthen the prominence of this therapeutic isotope. One goal is to establish an alternative production route for actinium-225 using less-problematic thorium-232 as the target material. On the other hand, efforts have been under way to develop target-tissue-efficient delivery systems for actinium-225 and its daughters. One very promising approach appears to be the use of water-soluble polymers to reduce daughter migration in vivo.

Neutron Capture Cross-Section Measurements Using the DANCE 4π Array

In a collaborative effort, scientists across organizations have been involved in an agile program to measure neutron capture and neutron-induced fission cross sections using isotopically pure actinide samples. The group has provided new absolute neutron cross-section data on a number of actinide isotopes.

The core of the Detector for Advanced Neutron Capture Experiments (DANCE) collaboration is composed of scientists from two Los Alamos groups: Nuclear and Radiochemistry (C-NR) and Neutron and Nuclear Science (LANSCE-NS). The larger collaboration also includes active participation from several other Los Alamos groups (Nuclear and Particle Physics, Astrophysics

This article was contributed by Todd Bredeweg, Marian Jandel, Robert Rundberg, and David Vieira of the Nuclear Radiochemistry Group; and Aaron Couture and John Ullmann of the Neutron and Nuclear Science Group.



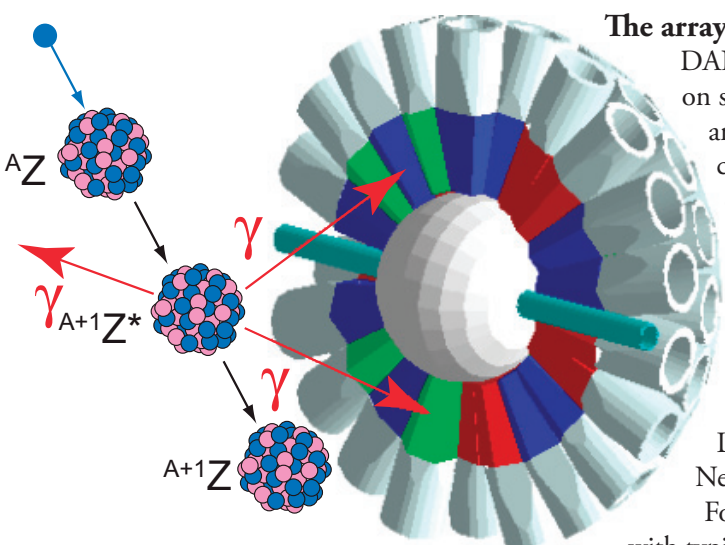
The DANCE detector is used to measure neutron capture and fission cross sections. Only half of the detector is shown.

and Cosmology; Applied Science and Methods Development; and Actinide Analytical Chemistry), other U.S. and foreign laboratories (Lawrence Livermore and Idaho National Laboratories, the French Atomic Energy Commission, and the Triangle Universities Nuclear Laboratory), and several universities (North Carolina State University, University of Nevada Las Vegas, University of California–Berkeley, and Colorado School of Mines).

Most of the recent work within the DANCE collaboration has focused on measuring neutron capture cross sections of interest to stockpile stewardship, attribution science, and the advanced fuel cycle initiative. Accurate neutron capture cross sections of many major and minor actinides are important for diagnostics of nuclear explosions and for accurate predictions of reactor performance, including buildup and burnup of minor actinides in the primary fuel.

Each of these programs requires a detailed knowledge of the neutron-induced reactions on a variety of actinide isotopes. Despite the fact that many cross-section data have been previously reported, the accuracy of the existing data remains an active debate. In particular, the uncertainties in the neutron capture to neutron-induced fission ratio ($\alpha = \sigma\gamma/\sigma f$) for many of the actinides are rather large, and some data are completely missing. These uncertainties are generally related to proper characterization and subtraction of background events in the experimental data.

The DANCE array was specifically designed to address these issues and provide the nuclear data needed by our sponsors. In support of these endeavors, the researchers have begun a program to produce high-precision cross-section measurements of the key production and destruction reactions of important nuclear fuel elements. The neutron capture targets that have already been fielded under this program include several isotopes of uranium, neptunium, plutonium, and americium.



GEANT4 model of one half of the DANCE array. The cartoon illustrates neutron capture on a target nucleus followed by emission of several gamma (γ) rays as the product nucleus de-excites to the ground state. The target sample is located at the center of the DANCE array providing nearly full solid-angle coverage (4π steradians), allowing detection of the full γ cascade.

The array

DANCE offers a unique capability to study nuclear reactions on small quantities of radioactive isotopes. The DANCE array is a 160-element barium fluoride (BaF_2) gamma-ray calorimeter with high granularity and nearly full solid-angle coverage (4π steradians), coupled to state-of-the-art data-acquisition electronics.

The primary purpose of the array is to perform neutron capture measurements on small (sub-milligram) and/or radioactive (activity = 1 curie or less, half-life = 100 days or longer) isotopically pure samples. The DANCE array is located on flightpath 14 of the Manuel Lujan Jr. Neutron Scattering Center at the Los Alamos Neutron Science Center (LANSCE).

Following neutron capture, the excited compound nucleus—with typical nuclear excitation energy of about 6 megaelectronvolts

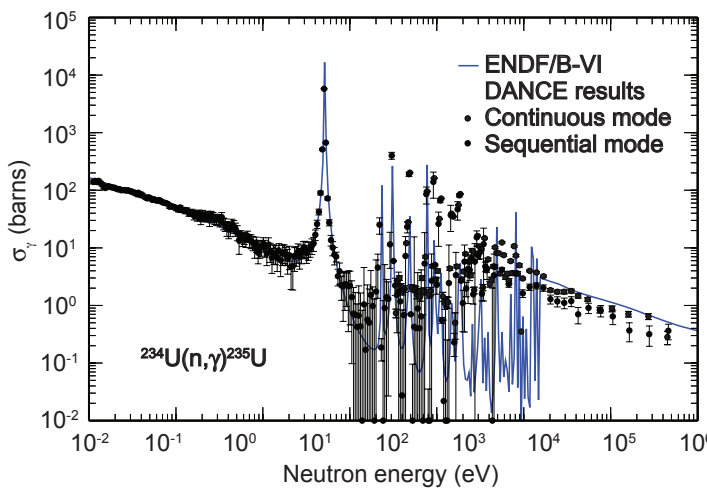
(MeV) for actinides—decays by emitting several gamma rays, de-exciting to the ground state or possibly to a long-lived isomeric state of the product nucleus. The DANCE array measures the emitted gamma rays, and by using the method of total gamma-ray calorimetry, researchers can identify the neutron capture events and extract the reaction cross section.

This represents a significant advancement over past measurements because the DANCE array measures the full energy of the gamma-ray cascade, which permits positive identification of the capture reaction and minimizes experimental backgrounds. The neutron time-of-flight method is used to tag each capture event with the incident neutron energy that produced the reaction. In this way, the neutron capture cross section is measured as a function of incident neutron energy.

A 6-centimeter-thick lithium hydride (LiH) shell, highly enriched in 6Li, surrounds the target/beam-line assembly to moderate and absorb a significant fraction of the neutrons that scatters from the target, preventing them from reaching the BaF_2 detector elements. This reduces one of the main background components, which consists of scattered neutrons captured on isotopes of barium in the detectors.

In spite of the power of the DANCE array to distinguish capture gamma rays from background, neutron capture measurements on many of the actinides are complicated by the presence of prompt gamma rays arising from low-energy neutron-induced fission, which competes with neutron capture to varying degrees. The addition of a dedicated fission-tagging detector to the DANCE array has greatly improved the ability to separate these two competing processes.

In addition to higher-quality neutron capture data, the addition of a fission-tagging detector offers a means to determine the capture-to-fission ratio in a single measurement, which reduces the effect of systematic uncertainties. Using a dual parallel-plate avalanche counter with the target material electro-deposited directly on the center cathode foil provides a high efficiency for detecting fission fragments and allows loading of preassembled target/detector assemblies into the neutron beam line at DANCE.



Uranium-234 neutron capture cross section for incident neutron energies 10 megaelectronvolt ≤ En ≤ 50 kilo-electronvolt. The solid line is the Evaluated Nuclear Data File (ENDF/B-VI) evaluated cross section, and the open and closed circles are DANCE results using two common data-acquisition modes. The DANCE data are normalized to the previously measured thermal neutron capture cross section. (A barn is a unit of area measurement of particles equal to 100 square femtometers; a femtometer is one quadrillionth of a meter.)

Recent results for uranium samples

The neutron capture cross sections of uranium isotopes are important for the accurate prediction of uranium fission reactor performance and for the use of uranium isotopes as a diagnostic for nuclear explosions. In spite of the importance of these cross sections, few measurements have been made, particularly at neutron energies around 10 kiloelectronvolts (keV) in part because of the difficulty of making these measurements.

Together with scientists from Oak Ridge National Laboratory, Los Alamos researchers measured the cross sections of uranium-234 and -236 and compared

them with previous data. In both cases the DANCE data were normalized to the thermal neutron cross section. The neutron capture cross sections determined for uranium-236 are in agreement with the previous ENDF/B-VI (Evaluated Nuclear Data File) evaluation.

However, the preliminary results for uranium-234 were lower than the ENDF/B-VI evaluation and are closer to older evaluations. The small bump at 0.3 electronvolt (eV) is probably due the (n,γ) reaction of uranium-235, a minor contaminant in the uranium-234 target. Results for both isotopes of uranium were used in the newer ENDF/B-VII evaluation released in December 2006.

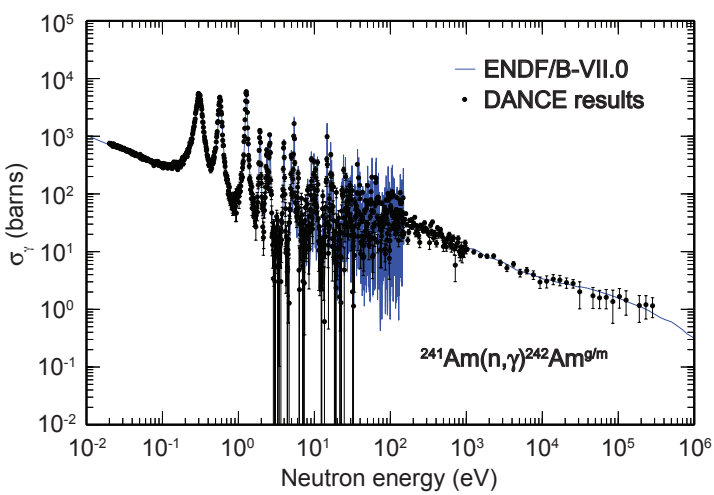
Recent results from americium samples

Scientists from C-NR and LANSCE-NS and external collaborators from Lawrence Livermore and Idaho National Laboratories have undertaken new measurements on the most prominent americium isotopes: americium-241, -242m (m = metastable), and -243. To properly use these isotopes as predictive tools in simulations, a precise knowledge of the cross sections for a variety of nuclear reactions [including (n,γ), (n,2n), and (n,fission)] on all of the americium isotopes is needed. These data are important for weapons diagnostics, understanding the burnup of americium in advanced nuclear reactors, and in nuclear forensics.

The first absolute neutron capture cross-section experiment on americium-241 has produced data for incident neutron energies between thermal (0.025 eV) and 320 keV, spanning seven orders of magnitude in neutron energy. The overall uncertainties in the measurement are typically 4 percent near thermal (0.025 eV), 6 percent near 1 keV, and 18 percent above 10 keV.

The SAMMY code, an R-matrix tool developed at Oak Ridge National Laboratory for fitting reaction cross-section data, was used to analyze the neutron capture resonances below 20 eV. The results of the SAMMY analysis indicated that the measured neutron widths show some significant deviations from previous data. The overall results of this measurement have the potential to significantly reduce the uncertainties in the radiochemical chain for americium.

Additionally, preliminary analysis has been performed for neutron capture on americium-243 at neutron energies between 10 and 250 keV, and use of the fission-tagging parallel plate avalanche counter in the center of the DANCE array has enabled characterization of the neutron-induced fission components for the neutron capture and neutron-induced fission cross-section measurements of americium-242m. The latter two data sets are currently being analyzed.



Americium-241 neutron capture cross section for incident neutron energies 20 megaelectronvolt ≤ En ≤ 320 kiloelectronvolt.

THE MATERIALS TEST STATION: A FUTURE IRRADIATION FACILITY

This article was contributed by Eric Pitcher of the Los Alamos Neutron Science Center.

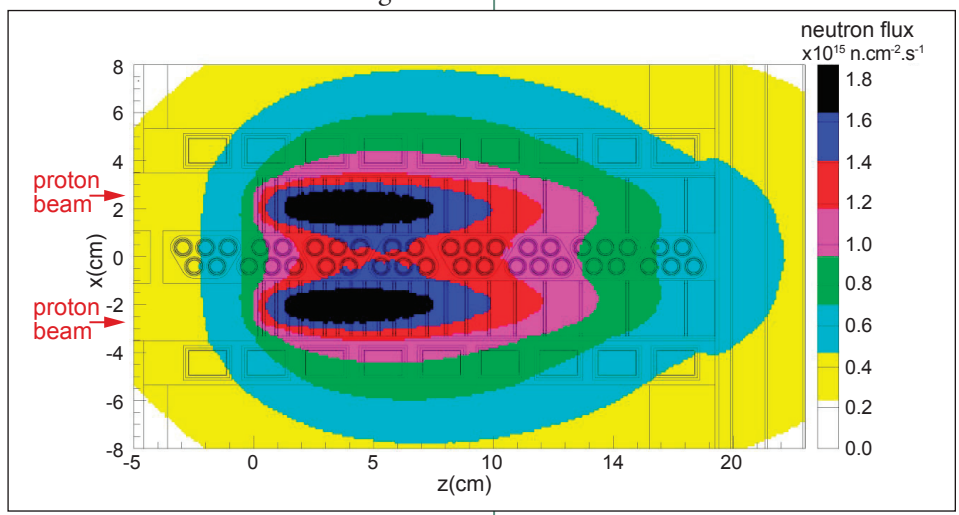
For nearly three decades, the Los Alamos Neutron Science Center (LANSCE) accelerator reliably delivered 1 milliampere (mA) of 800-megaelectronvolt (MeV) protons (H+) to pion-production targets in support of fundamental nuclear physics experiments. While the LANSCE accelerator has continued to deliver 100-MeV H+ beam to the Isotope Production Facility (IPF), the 1-mA proton beam has not been used since 2000 because there currently is no sponsored mission. Los Alamos and the DOE are planning to refurbish the LANSCE accelerator so that it can continue to reliably deliver beam for another two decades or more.

This refurbishment consists primarily of replacing antiquated power supplies that deliver radiofrequency power to the accelerator structure. Once refurbishment is completed in 2016, the LANSCE accelerator will be capable of delivering 1.25 mA of 800-MeV protons, or 1 megawatt (MW) of proton beam power, to the new Materials Test Station (MTS) with no reduction in beam delivery to all other existing facilities at LANSCE. The MTS will be located at the end of the accelerator in a former experimental area and is expected to be operational by 2015.

The basic configuration of the MTS target system consists of two spallation target regions separated by a fuel-irradiation region. The spallation targets are tall, narrow blocks of tungsten cooled by liquid lead–bismuth eutectic. The fuel irradiation region is 20 millimeters (mm) wide and can hold up to 40 fuel pins, each with an active fuel pellet stack height of 120 mm.

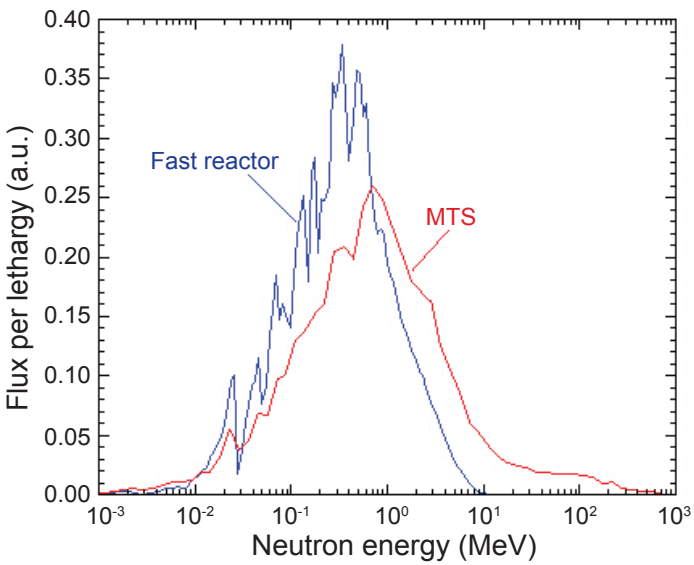
The pulsed nature of the proton beam allows alternate beam pulses to be directed onto one and then the other spallation target. Using two spallation target sections and alternating the beam pulses between them enables a nearly uniform time-averaged neutron flux in the fuel-irradiation region, which is located between the two spallation targets.

A spatial distribution of the neutron flux from a horizontal cut at target mid-plane shows the flux gradient across the 20-mm gap between spallation targets to be quite low. The peak flux in the fuel irradiation region is one-third to one-half of that found in the world's most intense research reactors.



A spatial distribution of the neutron flux from a horizontal cut at target mid-plane.

The MTS neutron spectrum at the peak flux position in the irradiation region is quite similar to that of a fast reactor, with the addition of a high-energy tail above 10 MeV that reaches up to the incident 800-MeV beam energy. The only significant impact of this high-energy tail is high hydrogen and helium production in metals. Compared to an equivalent flux level in a typical fast reactor, the peak flux position has about 60 times greater helium production rate in iron.



The neutron energy spectrum in the Materials Test Station compared to a typical fast-reactor spectrum.

For the ferritic/martensitic steels that are currently under consideration as fuel-clad material for fast reactors, high concentrations (greater than 600 atomic parts per million) of helium can lead to embrittlement. The impact of higher-than-prototypic fast-reactor helium production in candidate clad materials irradiated in the MTS must be given careful consideration with respect to assessing clad performance.

The spallation targets consist of tall (180 mm), narrow (21 mm) tungsten plates stacked along the direction of proton-beam propagation. The plates start out thin (4.4 mm) at the front end of the target and grow progressively thicker (up to 20 mm) toward the back. Between the plates are 1-mm-thick cooling channels. The plate thicknesses are adjusted in this manner so that the heat flux at the cooling surfaces is limited to 6 megawatts per square meter (MW/m^2), which does not challenge the thermal-hydraulic capability of the lead-bismuth coolant.

A critical parameter for obtaining meaningful irradiation data is the temperature at which the irradiations are performed. To achieve conditions similar to that of a fast reactor, the fuel-clad temperature must be controlled within a specified tolerance, and irradiation temperatures up to 550 degrees Celsius must be attainable in the MTS. These requirements are very difficult to achieve using water as the fuel coolant.

After considering a number of coolant options, lead-bismuth has been selected as the coolant for the test fuel pins. This coolant does not react exothermically with water or air (as is the case with sodium), has a high heat-transfer coefficient, and is liquid over a large temperature range. It has the disadvantages of not being a liquid at room temperature (requiring trace heating on loop piping and components), requiring active oxygen control to reduce corrosion, and producing polonium-210 as an activation product. Polonium-210 (half-life = 138 days) is an alpha emitter, and limiting its release during off-normal events will require special attention to the design of safety systems.

The MTS will provide a critical domestic irradiation capability to the DOE, allowing fast-spectrum irradiations of new minor-actinide-bearing fuels to start well before a new fast reactor can be expected to begin operation in the United States. The MTS will offer several opportunities for Los Alamos to produce an expanded portfolio of isotopes complementing the IPF. Targets could be placed in the intense 800-MeV proton beam reaching the MTS, as well as in the very high flux of spallation fast and slow neutrons that will be generated at the MTS.

Irradiation with 800-MeV protons would induce nuclear spallation reactions that produce neutron-rich isotopes, many of which cannot be produced in a reactor. Irradiation with neutrons at positions available at the MTS can be used to produce large amounts of isotopes, typically produced in reactors that are not available commercially.

End Notes

Changes in Leadership at the Seaborg Institute

David Clark, director of the Seaborg Institute, has been asked to lead the development of plutonium science strategy for Los Alamos National Laboratory. Seaborg Institute Deputy Director Gordon Jarvinen has assumed the director's position for the Institute in an acting capacity.

Los Alamos is the Plutonium Center of Excellence for the Department of Energy (DOE) complex and has under its mission the responsibility to lead science, engineering, and technology development across a broad range of plutonium-centric programs. Key Laboratory leaders have chartered development of an institutional plutonium science strategy to provide a road map for plutonium research and development efforts across the Lab's three mission pillars: nuclear deterrence, global threat reduction, and energy security.

In addition, the Seaborg Institute welcomes Wolfgang Runde as the new deputy director. Runde brings nearly two decades of experience in actinide chemistry to the Institute, with specialties in thermodynamics, separations, environmental, and radioisotope science. Runde is also the guest editor for this issue of *Actinide Research Quarterly*. For the past three years he has managed the production and distribution of reactor and accelerator isotopes across the DOE complex.

The Seaborg Institute would also like to welcome Joe Watts, who will continue his efforts as a program manager within the Stockpile Manufacturing and Support Directorate (ADSMS). Watts is also working to identify new program opportunities for actinide science within the DOE Offices of Environmental Management and Intelligence and participating in the Plutonium Science Strategy Working Group on Data Mining and Information Science.

David Clark

Wolfgang Runde

Gordon Jarvinen

Albert Migliori

Joe Watts

Seaborg Directors Named AAAS Fellows

Gordon Jarvinen and Albert Migliori have been named Fellows of the American Association for the Advancement of Science (AAAS). Jarvinen is acting director and Migliori is a deputy director of Los Alamos' Seaborg Institute for Transactinium Science and both are scientific advisors for *Actinide Research Quarterly*. They are the second and third members of the Seaborg Institute to be named AAAS Fellows; David Clark received the honor in 2005.

AAAS, founded in 1848, is the world's largest general scientific society and publisher of the peer-reviewed journal, *Science*. A Fellow is defined as "a member whose efforts on behalf of the advancement of science or its applications are scientifically or socially distinguished."

Jarvinen of the Stockpile Manufacturing and Support Directorate (ADSMS) was selected in the Chemistry Section for innovative research on processes that can be used to recycle plutonium and other actinides for national interests. His scientific accomplishments include research into water-soluble polymers and separation of plutonium-238 for NASA interplanetary space missions; the separation of trivalent actinides from lanthanides for advanced nuclear fuel cycles using liquid-liquid extraction; the fundamental coordination chemistry involved in extraction processes; and novel membrane separations that mimic ion-channels in biological systems.

Migliori of Condensed Matter and Magnet Science (MPA-CMMS) was selected in the Industrial Science and Technology Section for the development of resonant ultrasound spectroscopy and its application in materials physics and technology. Migliori, who came to Los Alamos as a Director's Postdoctoral Researcher in 1973 and became a staff member in 1976, is codiscoverer of acoustic heat engines and the leading expert in the use of resonant ultrasound spectroscopy.

Two other Los Alamos researchers were also named AAAS Fellows. John Sarrao of the Science Program Office (PADSTE-SPO) and Darryl L. Smith of Physics of Condensed Matter and Complex Systems (T-4) were both selected in the Physics Section.

ARQ wins technical communication award

Actinide Research Quarterly has earned a Distinguished Technical Communication award from the Society for Technical Communication (STC) East Tennessee Chapter in the technical publication competition. Three levels of recognition are available: Distinguished, Excellence, and Merit.

This is the first time *ARQ* has won a Distinguished; the publication previously has won eight Awards of Excellence and three Awards of Merit. As a result of winning at the highest level in a regional competition, *ARQ* will automatically be entered into the 2010 STC International Competition.

Three consecutive issues were submitted to the competition: "Plutonium Processing at Los Alamos," 3rd Quarter 2008; "Plutonium Futures The Science–Dijon, France," 4th Quarter 2008/ 1st Quarter 2009; and "History of the Seaborg Institute," 2nd Quarter 2009.

Highlights from the 33rd Annual Actinide Separations Conference

Lawrence Livermore National Laboratory hosted the 33rd Annual Actinide Separations Conference in May 2009 in Tahoe City, Calif. The conference brings together experts in the fields of chemistry, nuclear and chemical engineering, and actinide processing to present and discuss experiences, research results, testing and application of actinide separation processes. The exchange of information is critical to solving both national and international problems associated with the processing of nuclear materials used for both defense and energy purposes, as well as for the safe disposition of excess nuclear material.

Louis Schulte of Actinide Process Chemistry (PMT-2) was appointed Los Alamos National Laboratory's representative to the conference advisory board. Schulte replaces Elizabeth Bluhm of Pit Disposition and Precision Fabrication (PMT-10), who served on the board for several years. The 34th Annual Actinide Separations Conference will be hosted by Argonne National Laboratory May 18-21. A conference website including logistics information for this year's conference is at http://www.cse.anl.gov/2010actinide_conference/index.html. George Vandegrift of Argonne is the technical host this year and may be contacted at vandegrift@anl.gov or contact Schulte at (505) 665-1100 or lds@lanl.gov.

One of the highlights of the conference is the presentation of the Glenn T. Seaborg Award. The recipient is chosen by a panel of experts from DOE's national laboratories and is truly recognition from peers within the actinide separations community. The 2009 award was presented to Raymond G. Wymer.

Wymer worked at Oak Ridge National Laboratory in the Chemical Technology Division from 1953 until his retirement in 1991. During his years at Oak Ridge, he was involved in research and development in all aspects of the nuclear fuel cycle. Since his retirement, Wymer has consulted extensively in the areas of radioactive waste management, fuel cycle process chemistry, and site remediation for DOE and its contractors.

Seaborg, the award's namesake and first recipient, was the codiscoverer of nine actinide elements. *Actinide Research Quarterly* scientific advisor Gordon Jarvinen received the award in 2008. The deadline for nominations for the Glenn T. Seaborg Award this year is March 26.

Schulte provided *ARQ* with the following highlights of the conference:

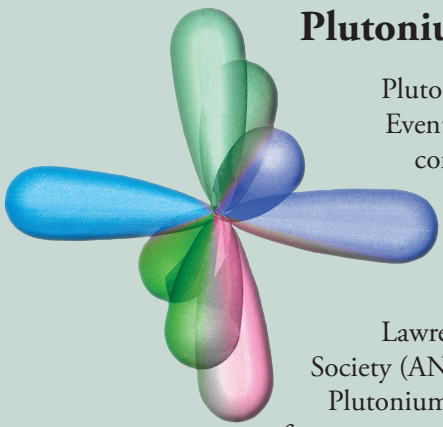
The 33rd annual conference drew participants from many DOE national laboratories as well as professors and students from the University of Nevada, Las Vegas; Washington State University; Oregon State University; and the University of California, Irvine. International participation came from the United Kingdom, France, Spain, and Japan. The conference is unique in that the central theme is not the same every year; host sites have the opportunity to focus upon particular areas of actinide separations. Thirty-five oral presentations were given over three days and the evening poster session included 15 presentations.

Plenary sessions focused on advanced fuel cycle actinide separations, global energy sustainability and nuclear energy, and actinide isotopic signatures with an emphasis on nuclear forensics. The demise of Yucca Mountain suggests that alternatives to permanent storage of once-through fuel are being considered, with $193 million allocated for fuel cycle research and development.

A significant area of focus at the conference was the Livermore site de-inventory efforts to achieve Security Category 3 status for "Superblock" (Building 332) as part of the DOE Complex Transformation. Security Category 3 will allow 400 grams of plutonium metal or 2000 grams of plutonium oxide (or a pro-rated combination). At this time 62 percent of Livermore's special nuclear material holdings have been packaged to DOE-STD-3013 and 55 percent of the material has been shipped to other sites.

Actinide solution chemistry, with emphasis on TALSPEAK actinide/lanthanide separations chemistry, was also discussed. Enough members of the "Sigma Team" (a cross-cutting team focused on practical design and process selection for separations in proposed U.S. nuclear fuel reprocessing efforts) were present to hold a breakout meeting separate from the scheduled conference activities. Actinide/lanthanide separations remain a very active area for development of new separations schemes.

Other topics of discussion included pyro- and aqueous process chemistry of actinide (actinium, thorium, uranium, plutonium, americium, and neptunium) separations at DOE sites, technetium-99 and actinium-225 separations efforts for medical isotope needs, nuclear forensics, fundamental studies on plutonium alloy metallurgy, and fabrication of nuclear fuel pellets. Talks were also given on processing of uranium ore, preparation of new reactor fuel, processing of spent fuel and fuel-cladding material, preparation and packaging of materials to DOE-STD-3013, surveillance and monitoring of those packages, and tank sludge stabilization/treatment and monitoring.



Plutonium Futures 2010 Set for September

Plutonium Futures—The Science 2010 will be held Sept. 19-23 at the Keystone Event and Conference Center in Keystone, Colorado. It will be the sixth conference in the series, which was initiated by Los Alamos and Lawrence Livermore National Laboratories in 1997. Previous conferences have been held in Santa Fe, New Mexico, in 1997 and 2000; Albuquerque, New Mexico, in 2003; Asilomar, California, in 2006; and Dijon, France, in 2008. The 2010 conference is cosponsored by Los Alamos and Lawrence Livermore National Laboratories and the American Nuclear Society (ANS).

Plutonium Futures conferences provide an international forum for the discussion of current research on the physical and chemical properties of plutonium and other actinide elements. By bringing people of diverse disciplines together, the conference aims to enhance the dialogue among scientists and engineers on the fundamental properties of plutonium and their technological consequences.

This year's conference will include discussions of condensed-matter physics; materials science; surface, interfaces, colloids, and corrosion; plutonium and actinide chemistry; fuel-cycle issues; and detection and speciation analysis. A panel discussion will focus on security issues of nuclear energy expansion. A special tutorial session will be held on the opening Sunday afternoon.

Pulitzer Prize-winning author Richard Rhodes will be the guest speaker at the conference banquet on Sept. 22. Rhodes is the author or editor of 23 books, including three volumes of nuclear history: *The Making of the Atomic Bomb*, *Dark Sun: The Making of the Hydrogen Bomb, and Arsenals of Folly: The Making of the Nuclear Arms Race.* His fourth and last volume of nuclear history, *The Twilight of the Bombs*, will be published just prior to the conference. Rhodes was the guest speaker at the first Plutonium Futures conference in 1997.

David Clark and Gordon Jarvinen of Los Alamos and Michael Fluss of Livermore are general chairs. David Hobart of Los Alamos' Actinide Analytical Chemistry Group (C-AAC) is program chair. Honorary chairs are Michael Anastasio, current Los Alamos National Laboratory director; Sig Hecker, Los Alamos director emeritus; and Gerry Lander, former director of the Institute for Transuranium Elements in Karlsruhe, Germany.

Information on registration, call for papers, and lodging will be available through the ANS webpage at http://www.new.ans.org/meetings/m_92. You may also link to the ANS web page through http://www.lanl.gov/conferences/pu2010/. Questions may be sent to Pu2010@lanl.gov; to Hobart at dhobart@lanl.gov, (505) 667-0205; or to Susan Ramsay at ramsay@lanl.gov, (505) 665-7214.

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sciences, atomic and molecular sciences,
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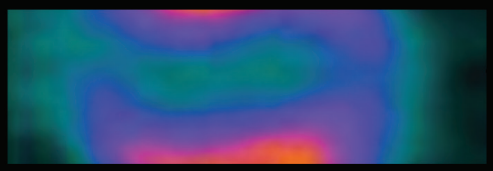

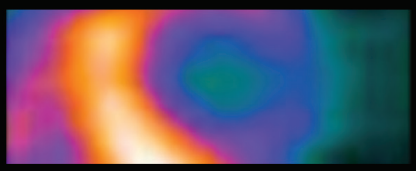

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