URANIUM-233 DISPOSITION, MEDICAL ISOTOPE PRODUCTION, AND BUILDING 3019 COMPLEX SHUTDOWN AT OAK RIDGE, TENNESSEE

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ABSTRACT

More than 1.5 tons of total uranium, including 450 kg of highly enriched ^{233}U, a special nuclear material contained in more than 1,200 canisters is stored in a Manhattan Project vintage facility. The Department of Energy (DOE) determined that continued ^{233}U storage in Building 3019A at the Oak Ridge National Laboratory represents a significant liability. At a minimum the material has to be repackaged to address safety issues and ultimately downblended to reduce security concerns.

The DOE concluded in their study of potential uses for ^{233}U, documented in *DOE Excess Material Disposition Decision Memorandum No. 2,* that the ^{233}U in Bldg 3019A is the only source of ^{225}Ac and ^{213}Bi in the Western hemisphere suitable for use in the treatment of cancer (e.g., clinical trials using the isotopes for treatment of acute myelogenous leukemia are underway at the Sloan Kettering Cancer Center in New York City, NY). The ^{233}U stored in the building was made from natural thorium in reactors because in the 1940s it was deemed potentially easier to chemically separate ^{233}U from thorium than to use enrichment to separate ^{235}U from ^{238}U. Between the 1940s and 1970 more than 2000 kg were produced for experimental use. Building 3019A has served as the national repository for ^{233}U since 1962.

DOE developed a project to (1) process and package the ^{233}U inventory in Building 3019A in order to eliminate the need for safeguards, security, and nuclear criticality controls and to render the material suitable for safe, long-term, economical storage, (2) extract ^{239}Th during ^{233}U processing to increase its availability for medical research and treatment, (3) operate the Building 3019 Complex during the ^{233}U processing and medical isotope production, and (4) place the Building 3019 complex in safe and stable shutdown status awaiting decontamination and decommissioning.

DOE initiated a competitive procurement to accomplish the project and awarded a contract in October 2003 to accomplish the full scope of work using a phased approach. The first phase of the project consists of equipment design and facility modifications. The second phase is to operate the facility, which includes the construction, installation and operation of processing equipment and conduct of the processing campaign. The third phase is to place the Building 3019 complex in safe and stable shutdown status.

Characteristics of ^{233}U, which contains varying levels of ^{232}U, factor into processing equipment design and include (1) a specific activity of 9.6 x 10-3 Ci/g, similar to that of ^{239}Pu; (2) a critical mass which is 1/4 to 1/3 of that of ^{235}U; and, (3) a ^{232}U daughter ^{208}Tl that emits a 2.6 MeV gamma-ray, resulting in radiation levels for a 1 ml sample of 2-4 R/hr at 1 cm, with some canisters emitting 300 R/hr. These characteristics resulted in designing processing equipment to be housed in heavily shielded hot cells with extensive contamination, radiation, criticality, and security controls. In addition, the variety of canisters to be processed presents a design challenge that was resolved by incorporating a mechanical cutter to cut open canisters remotely.

Processing design has been tailored to the material form and includes pre-sorting, calcining, as needed, dissolution in hot nitric acid, filtration, accumulation in safe tanks for accountability measurement, selective thorium extraction using ion exchange, uranium downblending with depleted uranyl nitrate, concentration, denitration and conversion to U_3O_8, and packaging of the oxide product for safe long-term storage. Thorium will be purified and packaged in 10 to100 mCi batches for shipment to isotope producers to extract ^{225}Ac to produce ^{213}Bi generators.

In order to provide safe, secure state-of-the-art facilities for processing, the building requires several modifications: (1) construction of a new heavily shielded 250-ton hot cell with manipulators for canister opening, pretreatment, and dissolution; (2) addition of equipment to an existing shielded cell for NMC&A measurement and downblending; (3) construction of a new heavily shielded hot cell with manipulators for thorium extraction, purification, packaging and storage; (4) demolishment and removal of existing equipment and adding equipment for denitration, U_3O_8 conversion, and product packaging; and, (5) installation of new analytical labs, a process off-gas scrubber, liquid and solid waste handling equipment and other support equipment.

In the process development and design phase, a number of key technical issues have been addressed that include: dissolution of uranium materials, thorium separation/purification and concentration, denitration technology selection, isotope partitioning and shielding, radon hold-up, product packaging, and security prior to and during processing.

The ultimate goal of this 8-year project is to process and downblend excess weapons-capable material to a safe and stable form in order to enhance safety and security, and to provide medical isotopes to support cancer research and treatment with the potential to provide material for as many as 6,000 cancer treatments a year.

INTRODUCTION

The ^{233}U Disposition, Medical Isotope Production and Building 3019 Complex Shutdown Project (the Project) has been developed by the Department of Energy (DOE) to meet four major objectives: (1) process and package the ^{233}U inventory in Building 3019A in order to eliminate the need for safeguards, security, and nuclear criticality controls and to render the material suitable for safe, long-term, economical storage; (2) extract ^{239}Th during ^{233}U processing to increase its availability for medical research and treatment; (3) operate the Building 3019 Complex during the ^{233}U processing and medical isotope production; and (4) place the Building 3019 complex in safe and stable shutdown status awaiting decontamination and decommissioning.

The Building 3019 Complex is a Manhattan Project vintage complex that has received numerous upgrades and modifications during its 60-year history. It is located at the Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee. Building 3019A, the main building, was originally constructed in 1943 as a chemical separations pilot plant for the Manhattan Project (see Figure 1). Between 1943 and 1976, Building 3019A functioned as a pilot plant for radiochemical reprocessing technology. In 1962, Building 3019A became the national repository for ^{233}U, which remains its current mission. However, long-term storage of the ^{233}U inventory in its current configuration represents a significant financial liability for DOE. Continued long-term storage in Building 3019A would require major capital upgrades and retrofits to critical facility systems that may not meet current standards.

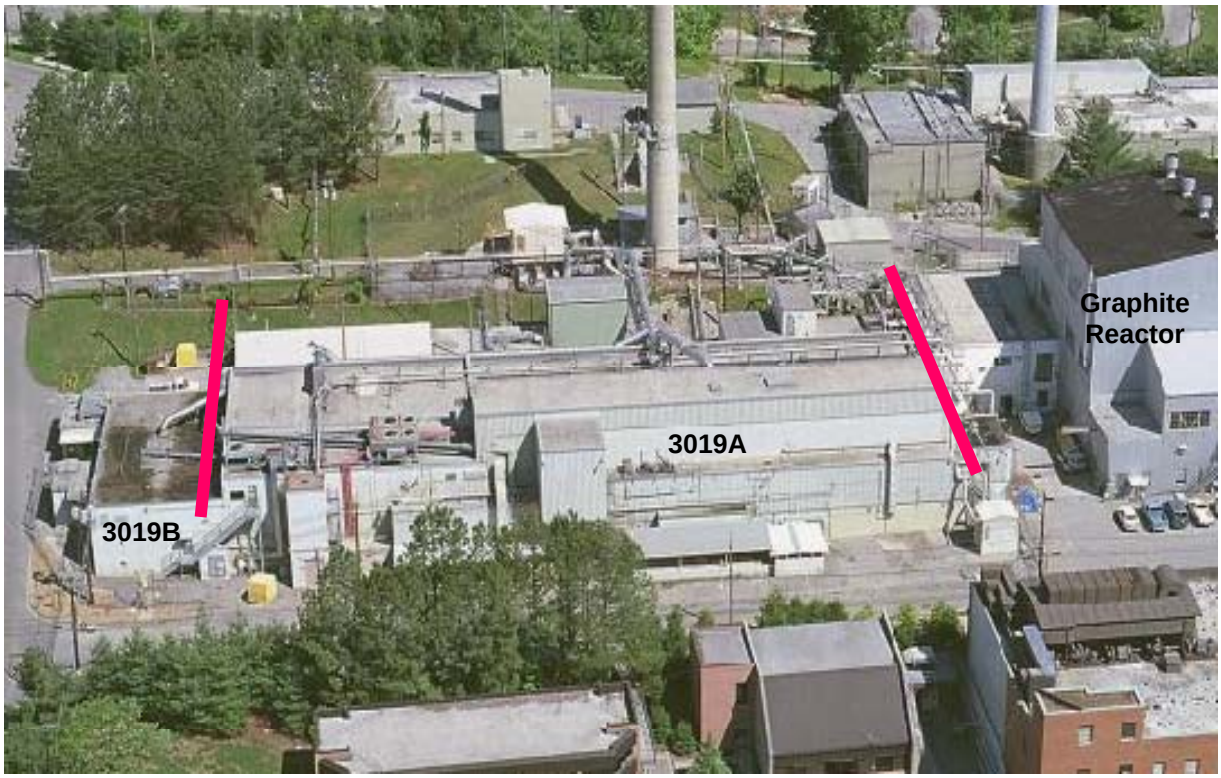


Fig. 1. Building 3019 Complex

The DOE has determined that there is no programmatic use for the ^{233}U currently in storage other than as a possible source of medical isotopes. The ORNL Building 3019A inventory of ^{233}U represents most of the readily available source of thorium-229 (^{229}Th) in the Western Hemisphere. Actinum-225 (^{225}Ac) and its progeny, bismuth-213 (^{213}Bi), are isotopes in the decay chain of $^{233}U/^{229}Th$ that have significant promise for cancer treatment and are in the research and clinical trial stage for treatment of acute myelogenous leukemia. These isotopes are also being explored for treatment of cancers of the lungs, pancreas, and kidneys.

The unique challenges presented by this project led to selection of an approach that entails ^{233}U processing within Building 3019A. Commercially proven technologies are being designed to fit within Building 3019A to convert the ^{233}U into a critically safe and stable storage form while simultaneously recovering the maximum amount of ^{229}Th.

Project Description and Organization

On March 3, 1997, the Defense Nuclear Facilities Safety Board (DNFSB) submitted Recommendation 97-1 to the DOE identifying the need to address the safety of near-term storage and the development of a long-term plan for uranium-bearing materials at ORNL. In response to the recommendation, the DOE issued an Implementation Plan on September 29, 1997 in which it committed, along with other near-term actions, to perform a study of alternatives to the continued use of Building 3019A as the National Repository for ^{233}U.

Since that time, the DOE considered the following alternatives:

- Possibility of alternate storage facilities within the DOE facility and laboratory network for ^{233}U handling and storage,
- Use of ^{233}U in thorium fuel cycle development,
- Use of ^{233}U as a calibration spike in determining uranium concentration and isotopic compositions in materials, or
- Continued long-term storage in Building 3019A.

None of the first three alternatives proved feasible. Continued safe storage of the ^{233}U in Building 3019A would require major facility and engineered systems upgrades. Storage of the inventory in its current configuration represents an approximately $15M per year liability for the DOE in order to meet the material-handling requirements and provide protection against nuclear criticality accidents or theft of the material.

On June 13, 2002, DOE issued a request for proposal (RFP) to procure a contractor for the disposition of the ^{233}U inventory. The RFP required the following scope of work:

- Processing and repackaging the ^{233}U inventory in Building 3019A to render it suitable for safe, long-term, economical storage including elimination of the need for criticality and safeguards and security controls,
- Extracting ^{229}Th from as much of the inventory as practicable in an effort to increase its availability to support cancer research and treatment,
- Operation of Building 3019A during contract performance, and finally
- Placing Building 3019A in a safe, stable shutdown mode.

As a result of the procurement process, DOE announced on October 9, 2003, that it would award the project, known as the ^{233}U Disposition, Medical Isotope Production, and Building 3019 Complex Shutdown Project, to Isotek Systems, LLC, which is a collaboration between Duratek, Inc., Nuclear Fuel Services, and Burns and Roe Enterprises. Additionally, DOE's Pacific Northwest National Laboratory will provide technical expertise in medical isotope separation and purification, and commercial medical isotope producers will provide medical isotope production and delivery.

Execution of the Project is broken up into three phases, as depicted in Figure 2. Phase I, which is the base contract award, encompasses preliminary planning, engineering, and design activities and DOE's NEPA evaluation. Phase II involves the facility modifications. At the beginning of Phase II simultaneous operations of the Building 3019 Complex and procurement and construction activities begin. Phase II approval will be contingent upon successful completion of Phase I. Phase III encompasses safe shutdown of the Building 3019A Complex. Decontamination and final decommissioning is not included in this phase.

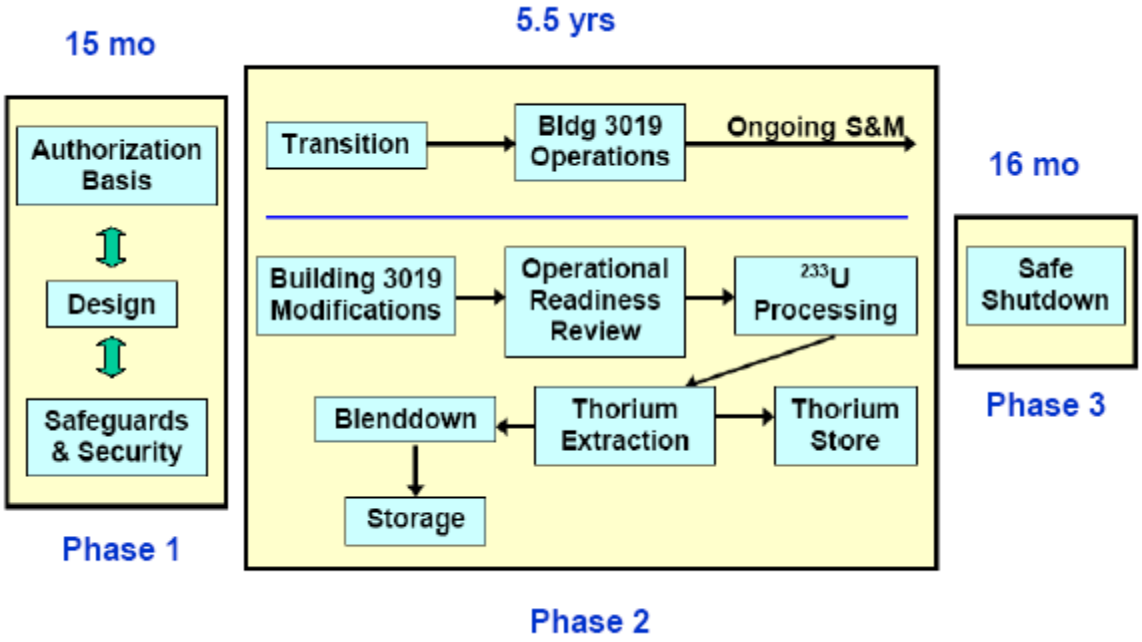


Fig. 2. Project Work Phases

History of Building 3019A

In early 1943, as part of the Manhattan Project, plans were made to build an air-cooled experimental graphite pile, a chemical separations pilot plant, and supporting laboratories on an isolated tract of land in Bethel Valley, Tennessee, known as X-10. These major installations became the prime function of the Clinton Engineer Works, now known as Oak Ridge National Laboratory. Since that time, Building 3019A has served as a pilot plant in the development of several radiochemical processes that have found application in both government and commercial facilities worldwide. In addition to the process development role, the facility's operations have also produced large quantities of plutonium, uranium of all isotopes, thorium, and special isotopes while processing irradiated fuel.

The Graphite Reactor, or Clinton Pile, was the first of its kind designed for the production of plutonium for use in nuclear weapons. The associated chemical reprocessing required was the extraction and purification of the plutonium from the reactor's spent natural uranium fuel. In 1943, several methods were proposed for this separation process. The first method selected was a precipitation process called the Bismuth Phosphate process, and it was used at ORNL from 1943 to 1945.

The first temporary buildings were started on February 2, 1943, and the buildings along with utility installations were completed in March 1943. Two months were required to complete the foundation for the cells in which the plutonium would be separated from the uranium fuel (see Figure 8). With concrete walls several feet thick, the cells would extend one story above ground and would be covered with massive, concrete slabs that could be removed to replace equipment. The first cell, linked to the Graphite Reactor building by an underground canal, contained a large tank in which the uranium fuel slugs and their aluminum jackets could be dissolved. The next four cells were designed for the equipment and piping for the successive oxidation-reduction cycles. The last cell served as a spare for storing contaminated equipment. Alongside the cells was a one-story frame building used for the operating gallery and offices. Cell wall construction was initiated in June.

The installation of piping and equipment began in September. The testing and extensive modification of process equipment required most of October, but the plant was ready to operate when the first fuel slugs were discharged from the pile in December. Photographs taken in 1943 of the construction of the Building 3019 are presented in Figures 3 and 4. The designation of the building at that time was Building 205.

Since this early beginning in 1943 and through 1976, Building 3019A was a pilot plant for numerous fuel recovery processes. This includes significant chemical processes such as Purex and Thorex (the Thorex process produced much of the ^{233}U now in storage in the building). The building complex in its current form is comprised of various annexes, support buildings, and irregular floor levels totaling a nominal 30,000 ft^2.

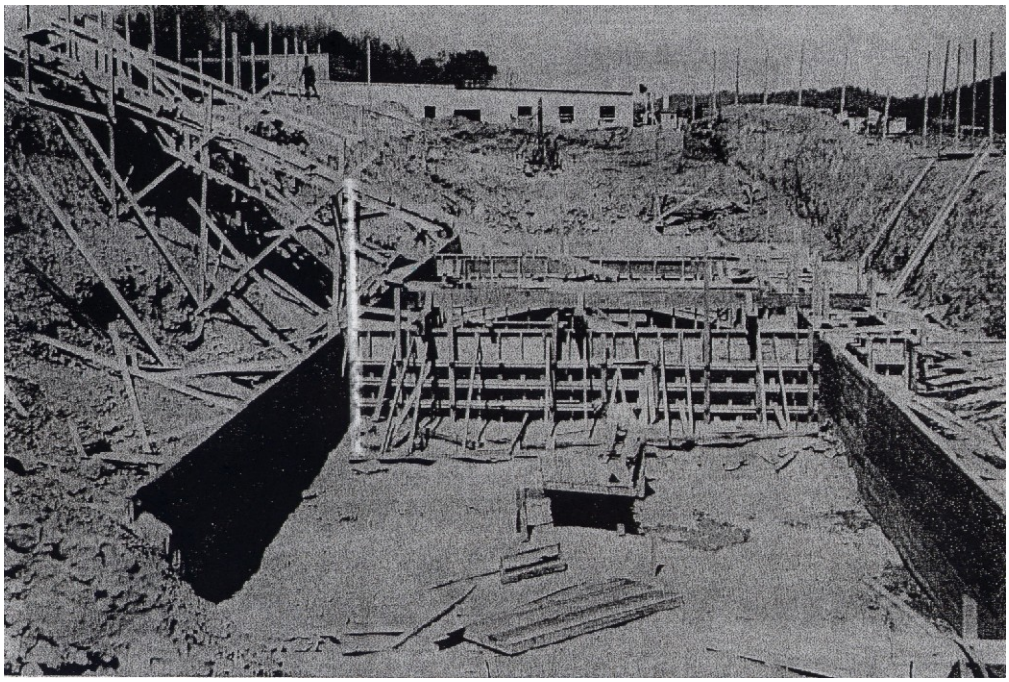


Fig. 3. Cell Foundation Excavation and Forms, May 1, 1943

Fig. 4. Original Cell Access at Ground Level, November 11, 1943

At the core of the complex is the original building structure, which consists of seven shielded processing cells positioned from east to west. Above the processing cell is a high-bay structure (the Penthouse). These cells are described in detail in subsequent sections. Building 3019A also contains operational laboratories with gloveboxes and hoods and several areas with out-of-service equipment.

History and Characteristics of ^{233}U

The United States produced a significant quantity of ^{233}U (about 2,000 kg) during the Cold War in support of national defense and other missions. ^{233}U is a man-made isotope that has been produced by irradiating natural thorium (^{232}Th) with neutrons. The ^{233}U produced is then chemically separated from the irradiated thorium targets.

In the production of ^{233}U, some uranium-232 (^{232}U) is produced. The concentrations of ^{232}U depend upon the specifics of the production techniques for ^{233}U. The ^{232}U has a decay product, thallium-208 (^{208}Tl), which decays to stable lead (^{208}Pb) and produces a 2.6-MeV gamma ray. Alpha radiation also presents a health hazard. The specific alpha activity of ^{233}U (9.6×10^{-3} Ci/g plus rapid buildup of short-lived alpha-decay products) is about three orders of magnitude greater than that for ^{235}U (2.2×10^{-6} Ci/g). High-energy alpha radioactivity necessitates glovebox handling for ^{233}U due to a phenomenon called alpha recoil. Alpha decay from ^{233}U tends to physically spread the radioactive decay products away from the original source.

The use of ^{233}U with the more complex thorium fuel cycle in the U.S. power cycle was ultimately abandoned in favor of lower low-enriched uranium (LEU) in the U.S. Some countries continue to investigate this option, however.

The storage and handling requirements for fissile materials must take into consideration containment, criticality control, safeguards, and shielding. ^{233}U has some similar properties to highly enriched uranium and weapons grade plutonium, as illustrated in Figure 5, but it also has its own unique properties that require heavily shielded storage and work areas with extensive contamination and criticality controls. Because of the high-energy gamma from the ^{208}Tl component, a 1 ml sample of the Building 3019A ^{233}U inventory measures about 2 to 4 R at 1 cm. Some canisters measure 300 R/h at 1 cm.

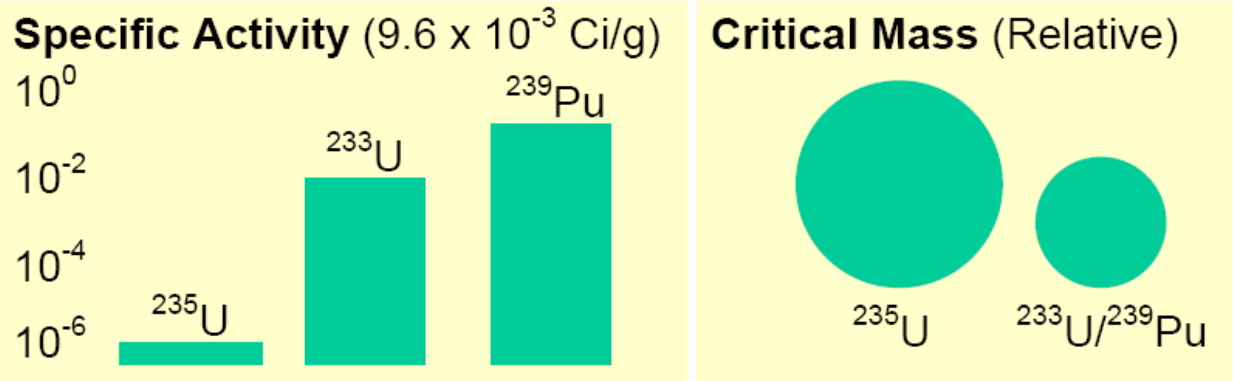


Fig. 5. Radiological Characteristics of ^{233}U

^{233}U Canister Inventory and Storage

Inventory

The inventory in Building 3019A consists of approximately 450 kg of ^{233}U contained in 1.5 metric tons of total uranium. There are approximately 40 g of ^{229}Th contained in this inventory and available for extraction. The inventory is primarily in the form of uranium oxides, but includes metals and other compounds. Uranium-232 (^{232}U) is present along with ^{233}U at concentrations ranging from 1 to about 220 parts per million (ppm).

The bulk of the material is contained in approximately 1,200 outer packages stored in shielded tube vaults within the building. In some instances, these outer packages contain multiple inner containers. Figure 6 shows some representative outer container types and Figure 7 shows two radiographs that are examples of inner containers and physical forms of ^{233}U. Of these 1,200 packages, some 403 packages and approximately 1,100 kg of the total inventory come from the Consolidated Edison Uranium Solidification Project (CEUSP). This material contains about 62-weight percent uranium (mixed ^{233}U and ^{235}U), and consists of an oxide monolith solidified into 24-inch long stainless steel containers (in Figure 6, the outer and inner CEUSP containers are the top two, respectively). This portion also contains relatively large amounts of ^{228}Th and its daughter product ^{208}Tl, which represent a significant radiation hazard. There are also 30 containers of similarly prepared, non-CEUSP material. Other large inventory groups include 140 containers of oxide powder and approximately 130 containers of U_3O_8, which has less than 10 ppm of ^{232}U. The group containing the largest number of individual items (1,700 pieces) is the unirradiated zero-power reactor fuel plates. Each plate is 2 by 3 by 0.25 inches and consists of $^{233}U_3O_8$ encapsulated in stainless steel. These plates are packaged into 130 tin-plate secondary containers (ZPR packets in Figure 7).

Based on visual and radiographic examinations of a sample of the canisters, review of available package records, and additional information available from sampling of the off-gas system that ventilates the storage tubes, there has not been a gross failure of any the packages. There is evidence of limited corrosion and pitting of the carbon steel storage tubes due to atmospheric moisture, but no evidence of condensate or accumulated water in the storage tubes.

The facility has also received ^{233}U for storage from the remediation of the Molten Salt Reactor Experiment (MSRE) at ORNL. In addition to the material stored within Building 3019A vaults, the contents of the P-24 Tank is also included in the processing scope of work. The P-24 Tank stores about 2,100 kg of natural thorium with a small quantity of ^{233}U in approximately 4,000 gal of thorium nitrate ($Th(NO_3)_4$) solution.

Fig. 6. Representative Outer Container Types Stored in Building 3019A Tube Vaults

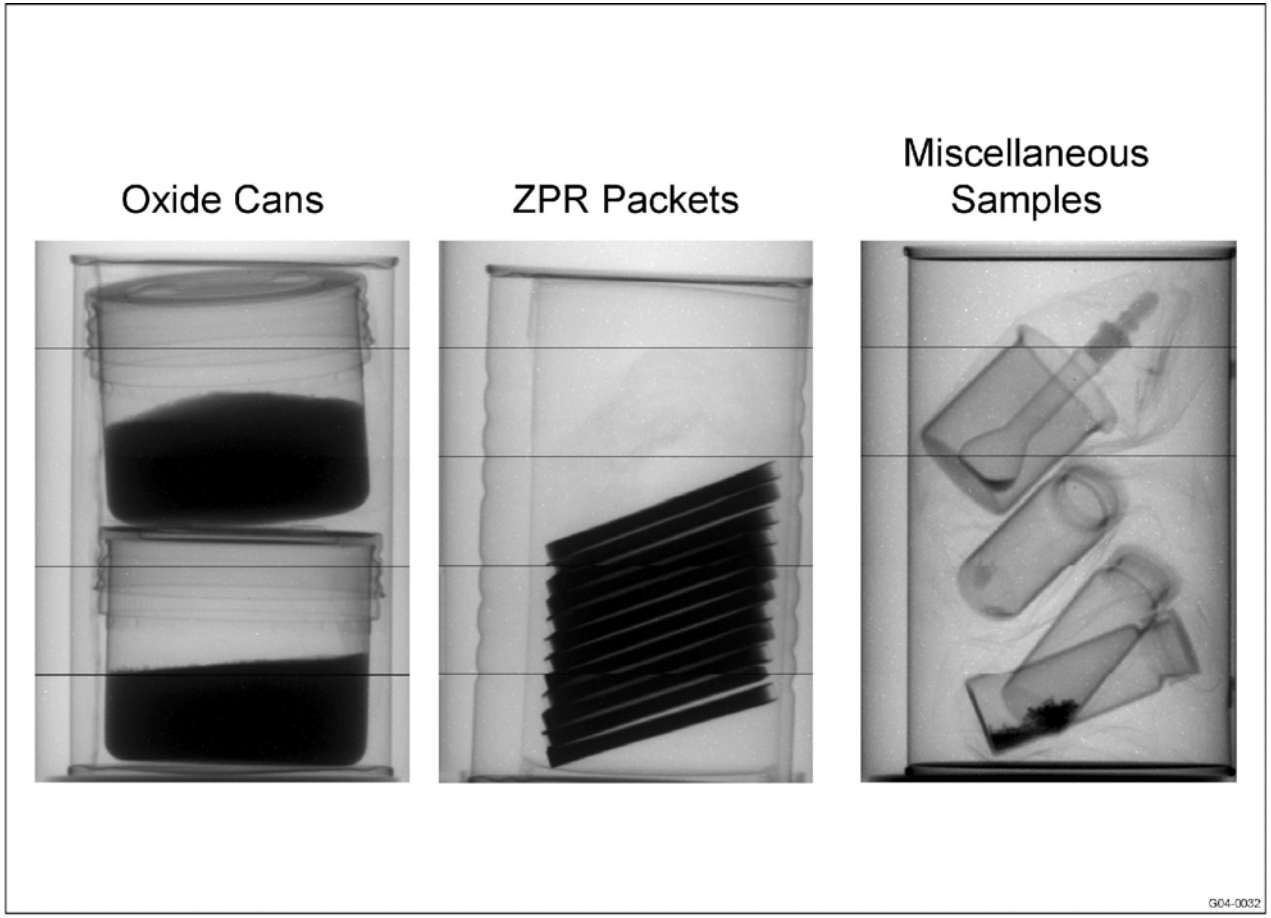


Fig. 7. Examples of ^{233}U Inventory

Storage

Building 3019A contains seven shielded processing cells (numbered as Cell 1 through 7) positioned from east to west (see Figure 8). All cell walls are built of poured, reinforced concrete. Each cell, except two, has a concrete roof hatch opening into the Penthouse.

Building 3019A contains four sets of top-loaded, shielded, storage tube vaults for the ^{233}U inventory. All tube vaults are top-loaded, shielded, ventilated, and accessible only under controlled conditions.

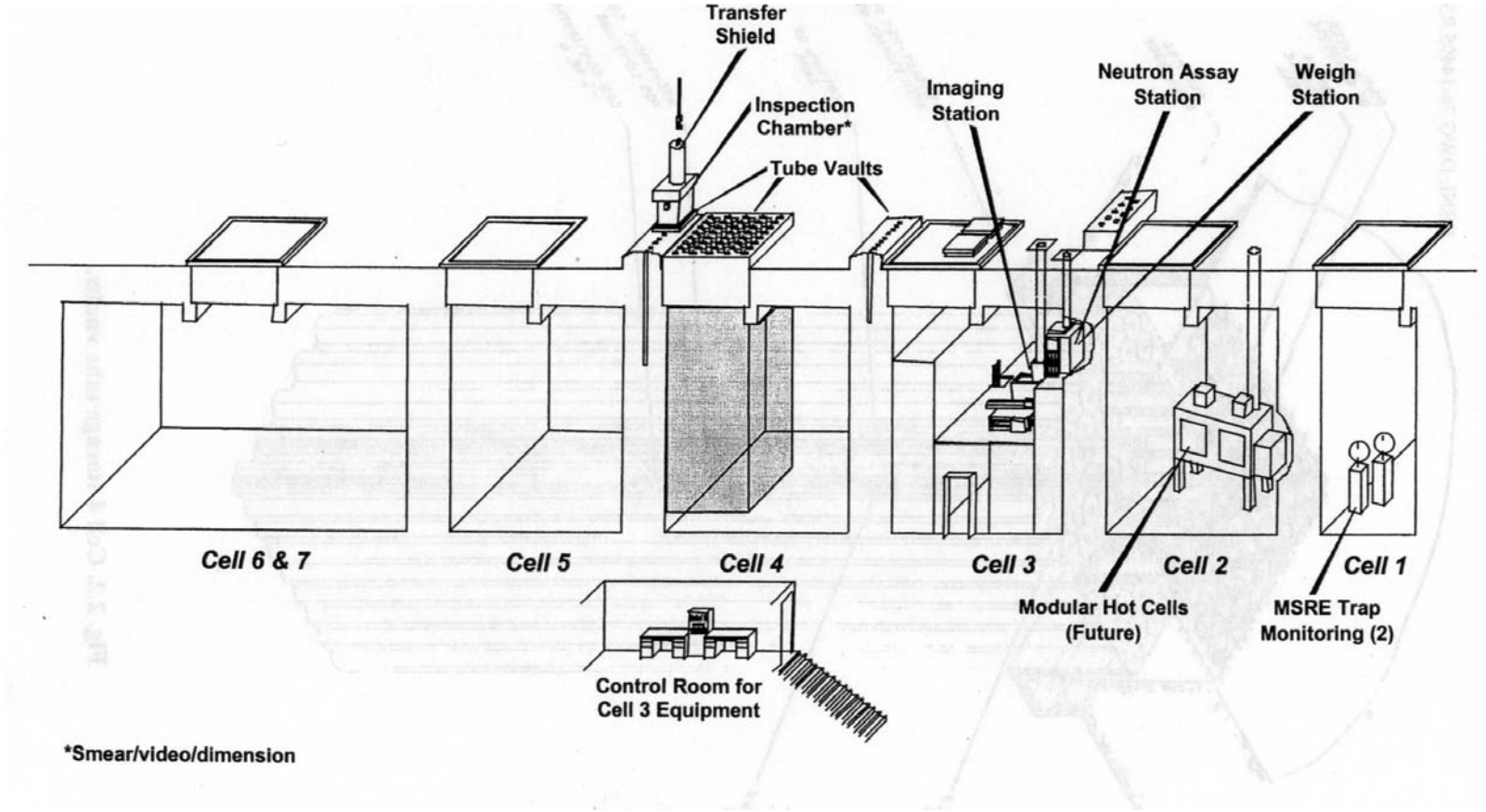


Fig. 8. Illustration of Building 3019A Cell and Tube Vault Configuration

Design and Process Description

Building 3019A will be modified to accept the new processing equipment necessary to support the scope of work. The overall process to extract thorium and downblend ^{233}U materials is shown in Figure 9.

Engineered Controls for Nuclear Safety

Due to the nuclear properties of the ^{233}U and ^{232}U all process equipment must be designed to ensure nuclear safety. Thus all pipes and tanks holding enriched uranium prior to downblending are limited to 4-inch diameter at a specified spacing and all tanks and sumps are limited to 1 inch in depth. Any exceptions will require stringent mass control.

Use of Hot Cells

Due to the nature of the ^{233}U material, the major processes will be conducted within hot cells. Hot cells provide two principal functions; confinement of radioactive contamination, and shielding from direct radiation. The common features of such structures include massive walls, thick viewing windows, remote operations with the aid of manipulators, and complex entry and egress ports for material throughput. The Project will employ the use of three hot cells as follows:

- Feed Material Preparations Hot Cell: Dedicated to accept, open, and pre-treat the wide variety of containers that are removed from the storage vaults.
- Thorium Hot Cell: Dedicated to the separation, purification, packaging, and storage of the ^{229}Th contained in the uranyl nitrate solution prepared in and transferred from the Feed Material Preparations Hot Cell.
- Denitrator Furnace Cell: Dedicated to the final processing steps of denitration and converting the downblended, thorium depleted uranium to U_3O_8.

Each hot cell is uniquely designed for its intended mission, and considers function, throughput, maintenance, mission life, and any future uses. A typical hot cell is shown in Figure 10. Descriptions of the hot cell features and processes within each cell are discussed in the following sections. Figure 11 shows a conceptual design for the Feed Materials Preparation Hot Cell.

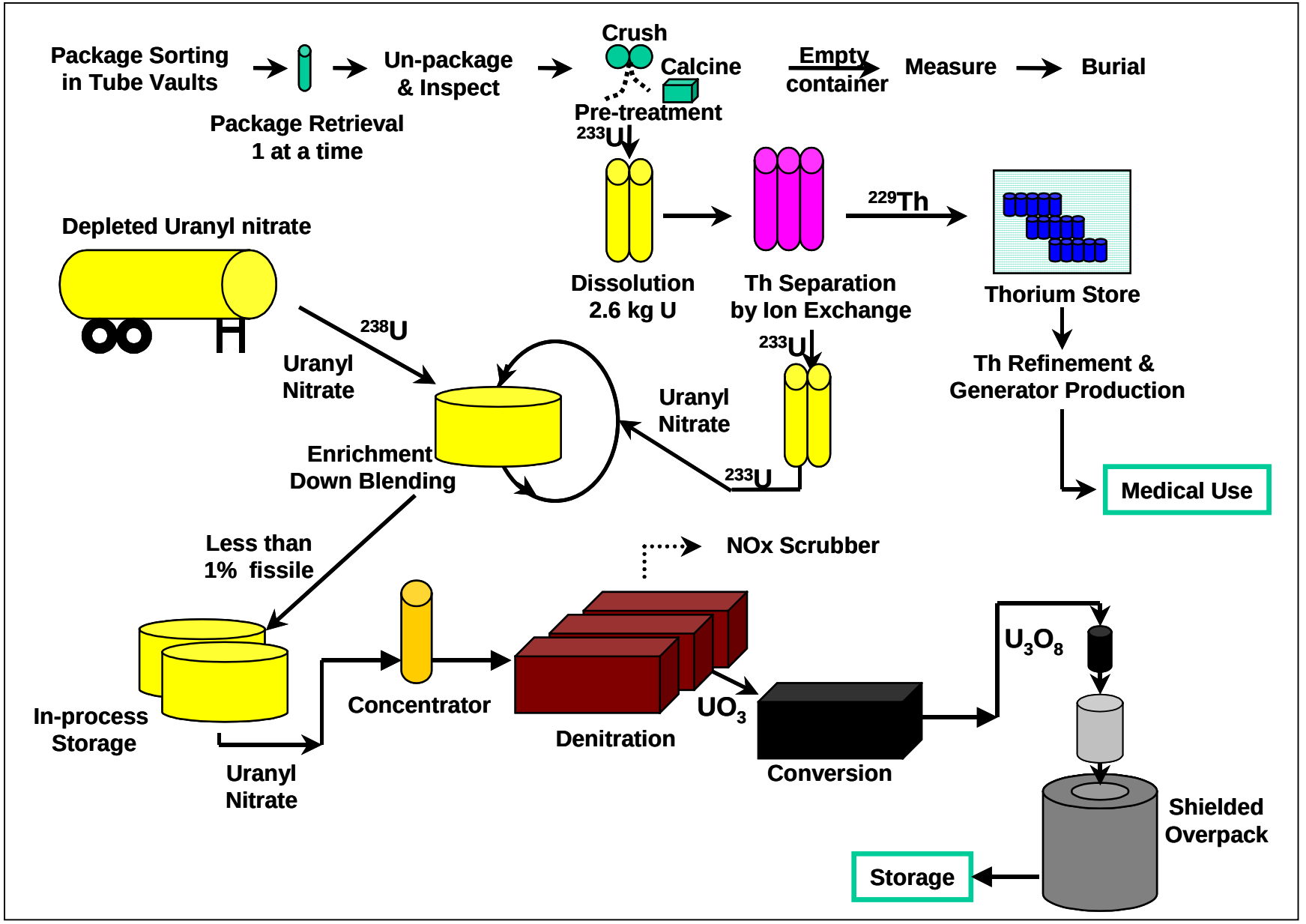


Fig. 9. Overall Process for Thorium Extraction and Downblending

Fig. 10. Typical Hot Cell

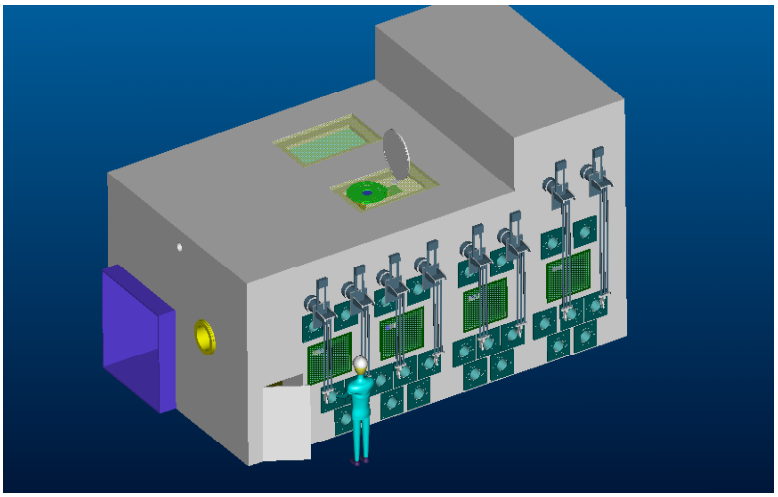


Fig. 11. Feed Material Preparations Hot Cell Concept

Pretreatment and Dissolution

Opening and Pretreatment

As a first step, the ^{233}U canisters will be retrieved, one at a time, and brought to the Feed Material Preparations Hot Cell where they will be inspected and weighed to verify they are the expected canister from the vault inventory. Once inspected, the canisters will be opened with the use of a mechanical cutter and any inner containers exposed and opened with the mechanical cutter or manipulators and specialized tools.

Inventory will be processed in two separate campaigns; CEUSP and non-CEUSP. The wide variety of uranium-bearing materials will require different pretreatment steps. Non-CEUSP material is present in various canister types. In each case the canisters will be opened with the use of the mechanical cutter. Any packaging materials present will be removed to a waste drum after inspection confirms that no significant ^{233}U material is present. Inner containers, which mainly contain U_3O_8 powder, will be transferred to the dissolver column station and the contents placed in the columns. The empty container will be vacuumed or rinsed with a weak nitric acid solution or water, with any rinse solution added to the dissolver. The empty container will be air-dried and internally inspected before placement inside a waste drum. A small percentage of the packages contain uranium metal foil and fluoride-bearing materials. These material will require calcining in a small furnace before proceeding to the next step, which is dissolution. The calcining step oxidizes metal material to U_3O_8, and removes residual fluoride and/or trace organics. Size reduction techniques (e.g., crushing) to enhance dissolution will also be used as necessary.

The CEUSP material represents about 75% of the total uranium inventory by mass. The CEUSP canisters are thick wall stainless steel with the contents solidified inside as a monolith. Rather than opening them with the mechanical cutter, the CEUSP canisters will be drilled down the long axis so that a core exists for the nitric acid to circulate through when they are placed into the dissolvers. The core-drilling residue will be collected placed into the dissolver.

Dissolution

After any necessary pretreatment, the uranium-bearing material will be placed into dissolver columns. An average of seven dissolution batches per week will be input. For the CEUSP material, this is typically one canister. The CEUSP container will be placed inside of a basket and then the basket into the column dissolver via a remote manipulator hook. Hot nitric acid will be down-flow circulated through the dissolver. The hook will then open the dissolver and remove the emptied basket/can. The can will be verified empty by either tare weight comparison to its stated tare weight or borescope inspection. Once the can has been determined to be sufficiently empty, it will go to a waste drum.

For the non-CEUSP material, multiple canisters will be added until the process batch size is achieved. Water and nitric acid will be added to the dissolver columns to form a highly-enriched uranyl nitrate solution. The dissolver columns will be heated, as required, to achieve efficient dissolution rates. The columns will also be mixed or agitated by recirculation pumps and an air sparge. Once dissolution is complete, the solution will be pumped through bag filters to a nuclear materials control and accountability (NMC&A) columns for measurements to verify material concentrations and quantities. The uranyl nitrate will be mixed within the columns. From the NMC&A columns, the uranyl nitrate will be transferred to the next Hot Cell for thorium extraction.

Thorium Extraction and Purification

Once the dissolution of the ^{233}U into uranyl nitrate is completed, the solution will be passed to the Thorium Hot Cell. This Cell is a remotely operated hot cell serving to extract, purify, and package ^{229}Th from HEUN generated in the dissolution processes. An identical system to extract ^{229}Th is provided for secondary streams produced in spills, flushing, and samples. A single thorium purification process serves both extraction processes (secondary and primary).

Extraction

In general, the HEUN solution produced in the dissolution step will be transferred from the UN storage columns to the Feed Storage/Acid Adjustment Column, where the nitric acid concentration in the feed column will be adjusted. A series of resin-filled ion-exchange columns will be used for primary ^{229}Th extraction, polishing, and eluting. Column duties will rotate as necessary to achieve thorium removal from the feed solution. After the thorium is removed the resulting HEUN solution will be collected in a storage column for downblending.

Thorium-bearing secondary streams will be processed in a near identical fashion as primary streams. Secondary streams will be concentrated in an evaporator and the nitric acid concentration in the feed column will be adjusted, as required. Once the resin is spent, it will be disposed.

Purification

The elutant from the thorium extraction ion-exchange processes (primary and secondary) will be concentrated by evaporation. The resulting solution will be passed through an ion-exchange column for thorium separation. The column will then be eluted with hydrochloric acid (HCl) to selectively remove thorium. The resulting solution will be diluted with HCl and re-concentrated by evaporation to ensure all the thorium is present in the chloride form. The resulting solution will be acid adjusted with HCl and passed through a final ion-exchange column to remove trace quantities of plutonium and uranium.

Packaging

Thorium product from purification step will be dispensed into vials. The product in the vial will be dried to a solid in an oven overnight. After drying, the vial will be capped, labeled for tracking purposes, and prepared for storage to await shipment. The vials would be placed in shielded containers and staged, prior to shipment to commercial customers, at a thorium storage area in hot cell space at Building 3019.

Downblending

The downblending process occurs after thorium separation. In the downblending process, the highly-enriched, thorium depleted uranyl nitrate (HEUN) from the Thorium Hot Cell is blended with depleted uranyl nitrate (DUN) to produce a non-fissile product that is equivalent to not more than 0.96 weight percent ^{235}U. The DUN solution will be produced by NFS using nitric acid to dissolve depleted UO_3 obtained from DOE's Savannah River Site.

A quantity of HEUN solution will be transferred from the measurement columns in the Thorium Extraction System to the HEU Feed Columns and then metered into the HEU Head Columns. The Down Blend tank will initially be filled to a specified level with DUN, which has been sampled and analyzed for gU/liter verification. The DUN solution will be transferred through a mass flow totalizer from the DUN Storage Tank located outside of Building 3019A. The Down Blend Tank level and density will be monitored to assure the correct quantity and concentration of DUN has been added to the tank. The correct quantity of DUN depends on the assay and isotopic properties of the HEU material. One of the two magnetic-drive, seal-less centrifugal recirculation pumps will be operated, and the DUN will be re-circulated through an eductor, where it will be mixed with a small stream of HEUN coming from a predetermined volume held in the HEU Head Columns. The eductor is utilized so that a HEUN addition to an empty Blend Tank cannot occur. A reduction orifice will be located on the HEUN feed line to limit the flow rate of HEUN. Recirculation and mixing will continue until all the HEU has been added to a batch.

After blending, a small amount of de-ionized water will be used to flush the HEU feed columns and feed pipe as required. After all HEUN and flush water has been added, the resulting solution will be mixed and sampled. Once the analysis is complete with an acceptable result, the downblended uranyl nitrate solution will be transferred to the Evaporator Feed Tanks.

Denitration and Packaging

The final step in the process is denitration and packaging. The down blended non-fissile uranyl nitrate solution is converted to a stable oxide (U_3O_8) through thermal denitration, where nitrogen oxide compounds and moisture and other volatile materials are driven off.

This Cell will contain process systems that will be required to convert down blended uranyl nitrate solution to U_3O_8. This is accomplished in several steps. First, the down blended uranyl nitrate solution stored in the Evaporator Head Tank is concentrated from approximately 485 to 1,000 g of uranium per liter in the Evaporator. The concentrated solution is then sent to the Denitrator Feed Tank. From this tank, the concentrated solution is fed to four Denitration Furnaces that remove residual water and convert the solution to solid UO_3 by heating it to approximately 350 ºC. The steam and NO_x from the thermal denitrification process is collected in a scrubber and off-gas collection system. The thermal denitration reaction is as follows:

$$UO_2(NO_3)_2 \cdot xH_2O_{(aq)} \rightarrow UO_{3(s)} + NO_{(g)} + NO_{2(g)} + O_{2(g)} + xH_2O_{(g)}$$

The Denitration Furnaces are horizontal, U-shaped, mechanically agitated trough furnaces. The uranyl nitrate solution is injected in the agitated bed, and quickly dries to form UO_3 granules. The UO_3 granules overflow out of the discharge end of the furnace.

Next, the UO_3 overflowing from the Denitration Furnaces is conveyed to a conversion furnace. In the conversion furnace, UO_3 is converted to U_3O_8 at approximately 800 ºC. The decomposition reaction is as follows:

$$3\ UO_{3(s)} \rightarrow U_3O_{8(s)} + \tfrac{1}{2}\ O_{2(g)}$$

The hot U_3O_8 product exiting the conversion furnace then flows into a product cooler. This non-contact cooler will use cooling water to remove heat from the U_3O_8 and cool it down to approximately 60 ºC.

The cool U_3O_8 granular product discharged from the product cooler will then be packaged for handling and storage in robust containers approved by DOE. These containers will then be transported to an ORNL Melton Valley storage facility and held for in-growth of thorium daughters for future extraction and medical isotope production.

Cancer Research and Alpha-Particle Radioimmunotherapy

A Brief History

Cancer is the second-ranking cause of death in the world today. In the United States, approximately 38 percent of the population contracts the disease and 17 percent succumbs to it. Today, physicists and physicians are working closely together to devise new methods for exploiting the power of ionizing radiation in the fight against this deadly disease. One such method, known as alpha-particle radioimmunotherapy, has become a promising strategy. In this method, monoclonal antibodies are used to deliver radioactive isotopes directly to tumor cells.

Radiation was first used to treat cancer around the turn of the century. In the 1920s, it was first used to cure laryngeal (voice box) cancer. This was a treatment breakthrough because surgery to treat this disease results in the loss of speech while radiation treatment can spare the ability to speak. During the ensuing 60 years, advances in both the radiation itself that made it possible to treat deep tumors and in the methods used to locate and evaluate tumors, brought about the evolution of radiation into a powerful form of cancer treatment. Today, about half of all cancer patients are treated with radiation.

Recent advances have made it even more useful. Radioactive implants allow delivery of radiation to localized areas with less injury to surrounding tissues than radiation from an external source that must pass through those tissues. Proton radiation also causes less injury to surrounding tissues than traditional photon radiation because it can be more tightly focused. Current research with radioimmunotherapy and neutron capture therapy provide ways to direct radiation exclusively to cancer cells, and in the case of radioimmunotherapy, to cancer cells that have spread to many sites throughout the body. Until the mid-1990s, the only way to treat

cancer that has spread to multiple locations throughout the body has been with chemotherapy, which uses drugs that preferentially kill dividing cells in a non-specific way.

Modern radioimmunotherapy, in which monoclonal antibodies are used to deliver radioactive isotopes directly to tumor cells, has been under development for two decades. Nearly all of this work has focused on the use of agents that carry beta particle–emitting isotopes. Beta particles have relatively low energy and long ranges, which make them most useful for radiosensitive tumors of considerable size, such as lymphomas. As an alternative, alpha-particle emitters such as actinium-225 (^{225}Ac) and bismuth-213 (^{213}Bi) are capable of extraordinarily potent single-cell kill of a wide variety of tumor types, including leukemias and lymphomas.

Unlike beta-particles, alpha-particles are extremely effective at killing cells through radioactive decay, and they deposit their energy over microscopic dimensions of only a few cell diameters. Therefore, antibodies "tagged" with alpha-emitters deliver a potent dose of radiation directly to the cancer with minimal or no exposure of healthy tissue. Phase II human clinical trials for treatment of a type of leukemia using ^{213}Bi are underway at New York City's Memorial Sloan-Kettering Cancer Center. ORNL is studying ^{213}Bi for lung cancer therapy, and the National Cancer Institute is conducting studies to determine the value of this therapy in treating various cancers as well as transplant conditioning.

Alpha–emitters are significant for the following reasons:

- *High local damage*. Alpha emitters compared to other radiation sources (x-ray, gamma, beta, etc.) deposit most of their energy in a very small volume within a few cell diameters. The large local energy deposition provides a higher assurance that the specific cell is destroyed, not just damaged. It is estimated that one to two ^{213}Bi decays will kill a cancer cell.
- *Auxiliary damage control*. In most types of radiation therapy, the radiation is concentrated on cancer cells, but healthy cells also receive high radiation doses. For example, if x-rays are used, many of the x-rays will be absorbed into healthy cells. Because alpha damage is very localized, secondary damage is minimized. This outcome is particularly important in treatment of certain cancers (e.g., leukemia) and other diseases (e.g., meningitis) where single cells or small clusters of cells are the targets that are interdispersed among healthy cells. Conventional radiation therapy will kill large numbers of healthy cells and have the potential to harm the patient.
- *Minimal long-term damage*. Most alpha emitters decay through many additional decays to a stable isotope. Each of these subsequent decays creates radiation damage beyond the cancerous cell that was destroyed. These longer-term effects can adversely impact the health of both patients and doctors. ^{213}Bi, however, has the desirable characteristic in that it and its decay products all have short half-lives and quickly decay after destroying the cancer. The half-life of ^{213}Bi is 46 min. It primarily decays by beta emission to ^{213}Po, which, in turn, decays to ^{209}Pb by alpha emission in 4×10^{-6} s. Both decays are simultaneous in terms of the destruction of cancer cells. The ^{209}Pb, with a half-life of 3.31 hours, decays in turn by low-energy beta emission to stable ^{209}Bi.

The ^{233}U decay chain is given in Figure 12.

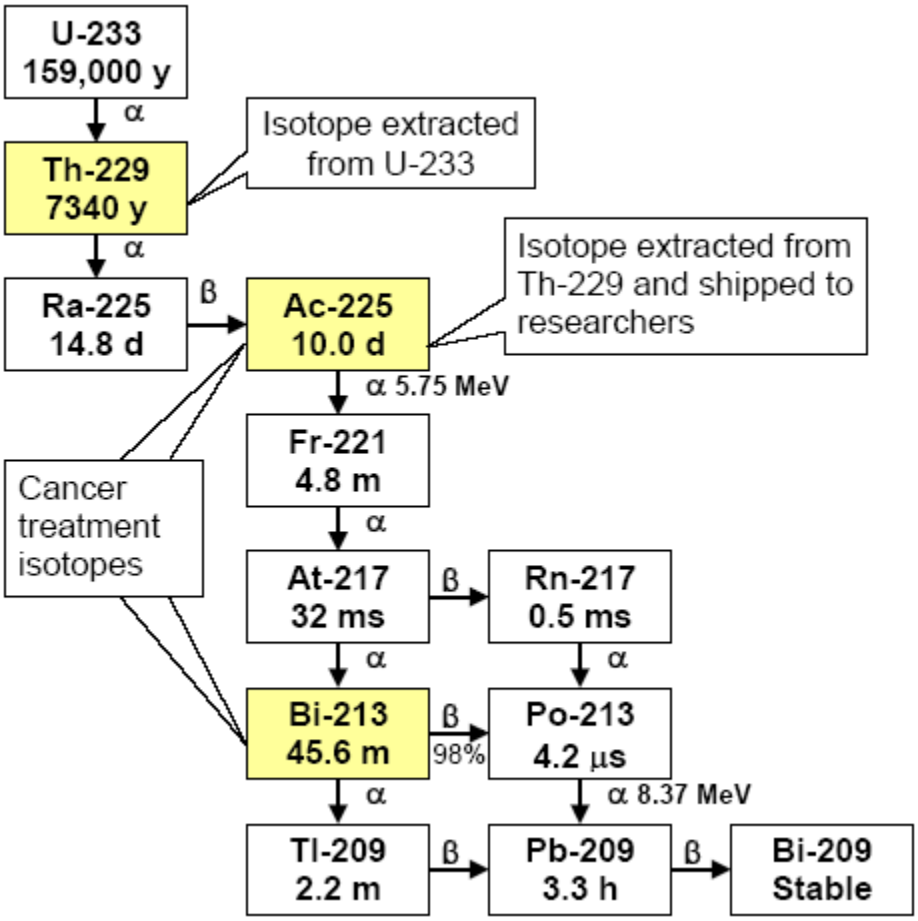


Fig. 12. ^{233}U Decay Chain

DOE's Contribution

For over 50 years, DOE has led the development of isotopes for medical diagnosis and treatment and for industrial uses. Examples of such developments are technetium-99m (used annually for about 85 percent of all diagnostic nuclear medicine imaging procedures worldwide), thallium-201 (for myocardial stress tests, iodine-123 (determination of thyroid disease), copper-67 (cancer therapy and imaging), tin-117m (promising agent for bone pain treatment and bone cancer therapy), and fluorine-18 (most sensitive radiotracer for detection and diagnosis of cancer). The DOE often partners with private industry by making its facilities and expertise available for the development of therapeutics. Each year, 600 deliveries of over 215 types of isotopes are made to over 300 domestic and international customers, including hospitals, pharmaceutical companies, and industrial customers.

With respect to ^{213}Bi, the DOE has been providing modest quantities of this radioisotope for the past several years. This work began in 1995 when the Nuclear Science and Technology Division of ORNL received funding to extract thorium from the sludge from the Building 3019A waste tanks. Waste from the original ^{233}U production during the Manhattan Project and Cold War days was stored in these tanks for 30 years. Fortunately, the extraction of the thorium proved relatively easy. Over that time, it was discovered that the thorium encrusted the boron-rich glass rings, called raschig rings, that had been placed in the waste tanks as a neutron absorber. Simply washing the rings in an acid bath retrieved much of the useful thorium.

After preliminary purification, thorium from the ^{233}U processing sludge was sent to a special hot cell in ORNL Building 3047 for further processing. The final product in the process, the ^{213}Bi, was then demonstrated by the Life Sciences Division to be an effective killer of mouse cancer cells. That work went on to show that ^{213}Bi treatment could cure mice of lung cancers.

In the past three years, ORNL's Isotope Program has been able to take advantage of inspections of the ^{233}U packaging to retrieve some additional ^{229}Th while some of the containers are being opened and inspected. Currently, the ^{229}Th is "milked" every 60 days to extract the ^{225}Ac. It is purified and sent to Memorial Sloan Kettering Cancer Center in New York and other institutions. At Sloan Kettering, about half a milliliter ^{225}Ac is loaded on a small column called a generator. The final product, ^{213}Bi, with a half-life of only 46 minutes, is eluted from the generator and incorporated into a monoclonal antibody, which is immediately injected into patients.

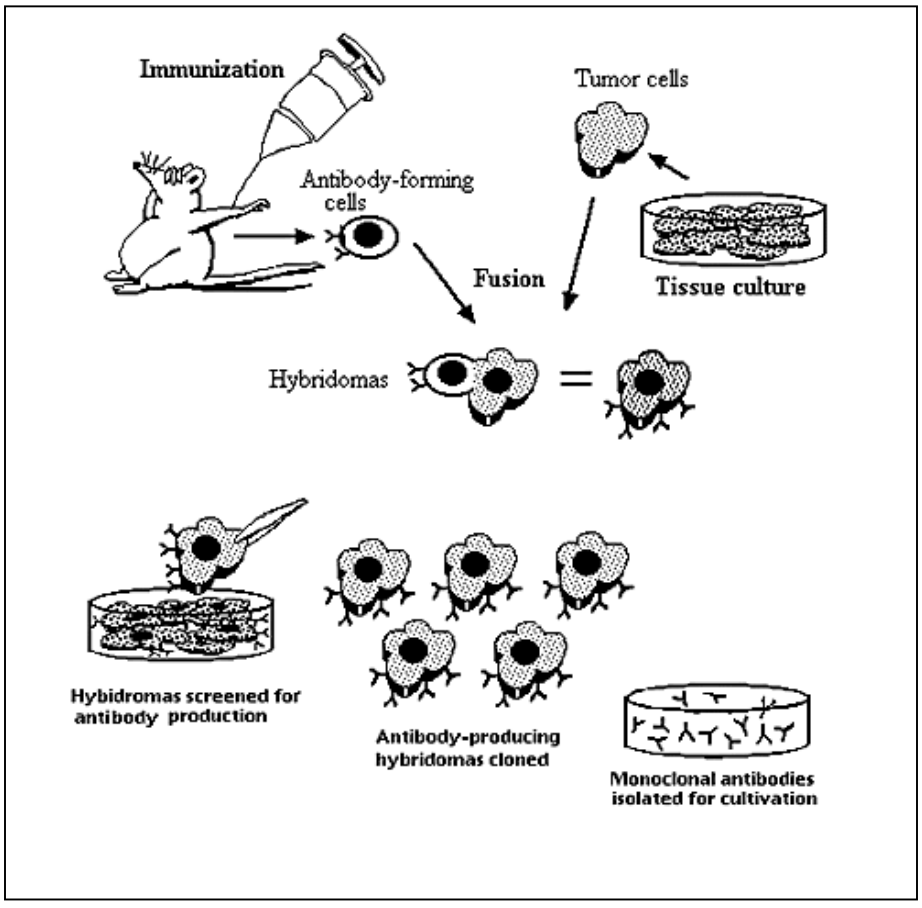
The 46-minute half-life of ^{213}Bi means it must be generated on demand and on-site in the clinical trials and for future therapy programs. The short half-life of ^{213}Bi fits well for the treatment of leukemia, however. The antibodies can find the leukemic cells in the blood within a few minutes. One or two alpha particles from ^{213}Bi are sufficient to kill the cell to which it is attached. Cells within a radius of about 100 micrometers (about 10 cell diameters) will also receive get a large radiation dose, but cells outside the 100-micrometer sphere will receive very little radiation. The treatment has been likened to a guided nuclear weapon on a cellular level, effectively performing the most precise of microsurgeries.
The quantities of ^{225}Ac now supplied by ORNL treat about 104 patients a year. With the U-233 Project, the number of patients treated per year is expected to increase by 5700% to 6000 per year.

Monoclonal Antibody Production

As mentioned above, antibodies are the vehicles that carry the radioisotopes. Antibodies are immune system molecules that specifically recognize and bind to only one molecular structure, and they can be designed to bind specifically to a certain type of cancer cell. To carry out radioimmunotherapy, antibodies with the ability to bind specifically to a patient's cancer cells will be attached to a radioisotope and injected into the patient's bloodstream. These special antibody molecules will travel around the body until they encounter a cancer cell, and then they will bind to it.

Human monoclonal antibody (MAb) production is quite complex. Simply put, the first step in MAb production is to immunize a mouse with an antigen (say, antigen X). This stimulates the

production of antibodies targeted against X, which are isolated from the mouse's spleen. The antibody-producing cells from the spleen are then fused with the cancerous cells grown in culture. The resulting cell is called a hybridoma. This product of cell fusion combines the desired qualities of both cells; the ability to grow continually, and the ability to produce large amounts of pure antibody molecules. By allowing the hybridoma to multiply in culture, it is possible to produce a population of cells, each of which produces identical antibody molecules. These antibodies are called monoclonal because they are produced by the identical offspring of a single, cloned antibody producing cell (the hybridoma cell). The process is shown graphically in Figure 13. Once a monoclonal antibody is made, it can be used as a probe to track down the specific protein that induced its formation. To effectively use the alpha-emitting isotope for cancer treatment, a stable linkage must be created between the isotope and the monoclonal antibody. This linkage is based on a chelation technology, and the linking molecule is called a chelator, as shown in Figure 14. Chelators are isotope specific, but generic for monoclonal antibodies.



Courtesy: Access Excellence

Fig. 13. Monoclonal Antibody Production

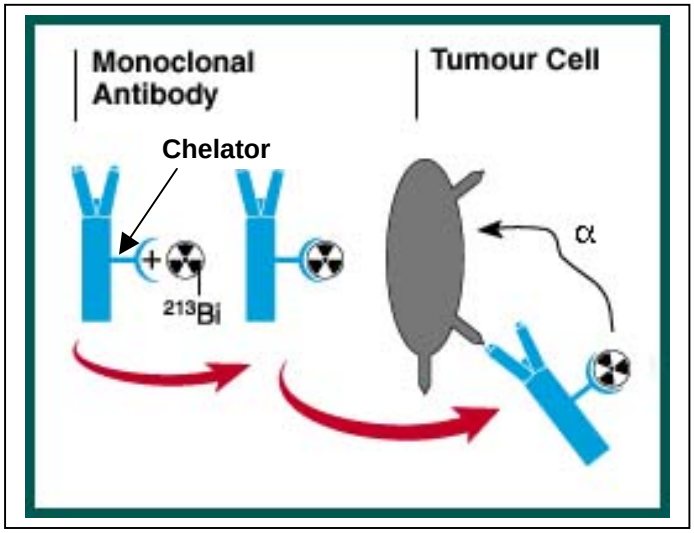


Fig. 14. Monoclonal Antibody

SUMMARY AND CONCLUSIONS

The continued safe storage of ^{233}U in the Manhattan Project-era Building 3019A represents a significant financial liability for the DOE. ^{233}U is a special nuclear material requiring stringent safeguards, security, and criticality controls. This expensive and unwanted nuclear legacy stored at ORNL will be downblended to a safe and stable form, and in the process, life-saving medical isotopes will be extracted.

There is a long history, experience, and significant record of accomplishments in DOE-supported radioisotope and radiopharmaceutical research, particularly at the national laboratories, going back over the last 30 years. This includes the promise of ^{213}Bi and alpha-particle radioimmunotherapy in fighting certain deadly cancers. The Project is not only important for its cancer treatment potential, but also for setting the stage for reducing global threats through the down-blending of materials "DOE has an important responsibility to clean up the dangerous materials and old contaminated structures left over from the Cold War," Secretary of Energy Spencer Abraham said. "That we can fulfill this mission while producing valuable new tools in the fight against cancer is an exciting and unique opportunity."

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