MODULE 5.0: ELECTROMAGNETIC SEPARATION (CALUTRON) AND THERMAL DIFFUSION

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Learning Objective

When you finish this section, you will be able to:

5.1.1 Describe the principles of the electromagnetic separation (calutron) enrichment process.

PRINCIPLES OF THE ELECTROMAGNETIC SEPARATION ENRICHMENT PROCESS

A large mass spectrometer called a calutron was developed by Ernest O. Lawrence at the University of California in 1942. An electromagnetic plant was built and operated at the Y-12 Plant in Oak Ridge, Tennessee, from late 1943 through the end of 1946 for uranium enrichment.

The world did not lack methods for separating isotopes when it discovered the possible utility of a kilogram of uranium-235 (U-235). Known techniques, pursued simultaneously in Germany and the United States, included ultra-centrifugation, diffusion across thermal or osmotic pressure barriers, and deflection in electric and magnetic fields. The last method appealed to Lawrence, who had made his reputation on the precise control of beams of charged particles. In principle the technique is simple. When passing between the poles of a magnet, a monoenergetic beam of ions of naturally occurring uranium splits into several streams according to their momentum, one per isotope, each characterized by a particular radius of curvature. Collecting cups at the ends of the semicircular trajectories catch the homogenous streams.

Note: The diameter of an ion's trajectory depends on the strength of the magnetic field and the mass, electrical charge, and speed of the ion. Lighter ions are more easily deflected by the magnetic field and consequently have a smaller diameter trajectory. For the separation of U-235 and U-238, the diameter of the 235 U⁺ ion beam is about 0.6% smaller than that of the ²³⁸U⁺ ion. In an electromagnetic isotope separator with a beam trajectory diameter of four feet, the difference in beam trajectory diameters for the 235 U⁺ and 238 U⁺ is about 0.3 inches. To properly focus the ion beams on the collector slits, the voltage of the ion accelerators and the electrical current in the magnets must be controlled to better than $0.1%$.

Basically, electromagnetic separation methods use magnetic and electronic forces to manipulate charged isotopic species. In this process uranium tetrachloride (UCI4), a uranium chloride salt. is electrically heated to produce UCI₄ vapor in an evacuated tank and ionized to give each molecule an electrical charge. These ions (uranium metal and chlorides) are passed through electrically charged slits to create a beam of charged particles that is passed

through a very strong magnetic field. The magnetic field introduces a force on each ion causing it to travel in a circular path, the radius of which is proportional to the momentum of the ion. The heavy U-238 metal ions have a greater momentum than the U-235 metal ions and their beam is bent less than the lighter ions. With this very strong magnetic field there is sufficient beam separation to permit individual collectors to be located to accumulate the light and heavy isotopes. Figure 5-1 shows a simplified schematic diagram of a calutron. At the Oak Ridge Y-12 plant, the separators were referred to as calutrons because development work on these separators were carried out using cyclotrons at the University of California-Berkeley. Some calutrons had two ion source beams, while others had four.

Over the years,

calutrons have been regarded—erroneously some feel—as an anachronistic technology. Essentially a scaled-up version of a mass spectrograph, they are slow and costly to operate. After World War II, they were replaced as the mainstay process for uranium-235 separation by less expensive technologies such as the gaseous diffusion process. Nevertheless, the Y-12 electromagnetic plant during World War II had 1,152 calutrons that were operated mostly by women who worked in shifts covering 24 hours a day.

100 milliampere source and all operating within the 184-inch field, would produce four grams of enriched uranium a day. The "S-1" committee that oversaw the uranium project for the Office of Scientific Research and Development (OSRD) recommended expending \$12 million to create a plant with 25 times that capacity before the fall of 1943. Lawrence did not doubt that other means, particularly reactor production of fissile plutonium, might ultimately be the most efficient way to a bomb. But in mid-1942 no reactor worked, and the calutron did.

Self-Check Questions 5-1

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

1. Describe the principle of the electromagnetic separation (calutron) uranium enrichment process.

- 2. What are the advantages of calutrons?
- 3. Which responds more readily to a magnetic field, light or heavy ions?
- 4. What were some of the technical problems associated with the electromagnetic process?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.2 Describe general facility and component layout of the electromagnetic separation enrichment process.

FACILITY DESCRIPTION AND COMPONENTS

of uranium in the beam. Once good beam conditions are established and the beams are going into the collector, only occasional adjustments are necessary. In a calutron with several ion sources, however, the failure of one beam can cause all the beams to fail.

The large vacuum chamber is situated between the pole faces of the electromagnet. "Forepumps" are used to begin pressure reduction; vacuum is maintained primarily by one or two high capacity diffusion pumps with pipe-throat diameters of 15–20 inches.

Special disposable stainless steel, water-cooled liners are often used in the vacuum chamber to simplify recovery of the large amounts of uranium that end up on the chamber surfaces.

A calutron electromagnet has two circular poles, separated by a gap 30–60 centimeters wide in which the vacuum chamber is inserted. The magnet is typically about one to two meters in diameter, weighs about 10–20 tons, and contains about a quarter-mile of thick copper wire. These extremely powerful magnets use one-third to one-half of the energy consumed by calutrons, and require cooling.

The power supply for the magnets requires a direct-current capacity of about 1,000 amps at 300–800 volts, similar to that used to power elevators. But the ones for calutron magnets must also be regulated precisely to produce a stable magnetic field.

The associated chemical processing area is usually one of the largest parts of the plant. In this area, the collectors are burned and the uranium is recovered from the ashes. In addition, the waste uranium must be recovered from the other calutron components by scrubbing with nitric acid, soaking in acid baths, or burning or dissolving away disposable components. The power supplies, magnets, and other components require extensive cooling with oil or purified water.

In practice at Y-12 the uranium enrichment was produced in two stages. The first, called ALPHA, was fed 0.711% normal assay $UCI₄$ and some 0.89% from the thermal diffusion process. The product from the ALPHA stage, along with the early production from the gaseous diffusion process, was fed into the BETA stage, which was able to enrich the material to above 90% U-235.

The ALPHA separator tanks were about 12 feet by 8 feet by 28 inches and weighed about 10 tons. The BETA separator tanks were about 6.5 feet by 5 feet by 23 inches and weighed about 9 tons. Magnets for the separators weighed on the order of 10 tons. Large overhead cranes were needed to transport these units within a building.

At the Y-12 plant, calutrons were not operated as single units, but were arranged in continuous oval or rectangular arrays called race tracks or simply, tracks. In these arrangements, separator tanks alternated with electromagnetic units. The track configuration allowed more efficient use of magnets and floor space. The tracks contained in close proximity the separators, magnets, and vacuum pumps.

The original calutron consisted of a large magnet, 4.67 meters in diameter. This magnet consisted of two circular pole faces separated by a gap in which a vacuum chamber was inserted. Inside this chamber could be placed a source of uranium ions (UCI_A) and, 180° around the circle, a collector. The source would accelerate the ions to a velocity such that the curvature of their trajectory in the magnetic field would cause them to arrive at the detector. The major problems encountered in the Y-12 calutron project were the severe limitations on ion beam strength created by space charge effects, and the inability to convert more than a small fraction of the feed material into product in a single run. Figure 5-2 shows one of the units developed during the Manhattan Project days. Figure 5-3 is a calutron unit diagram. Figure 5-4 shows a large number of the units installed at the Y-12 Plant in Oak Ridge. Y-12 featured nine first-stage ALPHA race tracks and four secondstage BETA race tracks. The ALPHA 1 calutron enriched uranium electromagnetically, in a collection of semicircular massspectrometer chambers (spares in left foreground), mounted side by side between powerful electromagnets (the tops of their rectangular frames are visible).

Figure 5-2. Calutron Unit

Figure 5-4. ALPHA 1

The calutron design settled on in 1942, called "ALPHA," provided for enrichment of natural uranium to about 15 percent U-235. Extravagant effort went into designing powerful ion sources and aptly shaped, eventually parabolic collecting slots. The many modifications and security codes proliferated whimsical names: sources Plato, Cyclops, Bicyclops, and Goofy mated with receivers Gloria, Irene, Mona, or Zulu. Ions from Plato and his friends traversed an arc 48 inches in radius to reach collector slits placed 0.6 inch apart. The guiding magnetic field was shimmed in obedience to calculations. Accurately machined and installed, the shims greatly increased the usable beam that reached the collectors. The magnet shims also increased the output of uranium-235. See Figure 5-5, installing magnet shims.

Figure 5-5. Installing Magnet Shims in an Alpha Calutron Tank to Increase Output of Uranium-235

Among results obtained with the 184-inch magnet was a design superior to it for large-scale calutrons, the so-called "XA." The prototype of the magnets to be installed at Oak Ridge, XA was a rectangular, three-coil magnet giving a horizontal field in which the calutron tanks could stand side by side. It had room for four ALPHA tanks, each with a double source. By the spring of 1943, convinced that the Germans might be ahead, Leslie R. Groves decided to skip the scheduled pilot plant; from the XA and a scale model of the production magnet alone would come procedures for ALPHA operation at Oak Ridge. Tests of the first, full-scale system installed there, the XAX, were scheduled for July. Figure 5-6 shows Frank Openheimer and Robert Thornton examining a 4-source emitter for the improved ALPHA calutron.

Figure 5-6. Frank Oppenheimer (Center Right) and Robert Thornton (Right) Examine the 4-source Emitter for the Improved Alpha Calutron

The spring and early summer of 1943 brought hundreds of trainees to Berkeley from Tennessee-Eastman Company, the operator for the Oak Ridge plant. The Laboratory labored to ensure that the test XA magnet system and ALPHA units were working by April in spite of delays in delivery of steel. Between April and July the training sessions ran continuously. In June a migration that by 1944 would reach 200 started for Oak Ridge. Laboratory expenditures exceeded half a million dollars a month.

The first wave of Berkeley workers at Oak Ridge had to see that the XAX magnet worked. Then runs could begin on the first production system, or "race track;" a 24-fold magnification of the XA that could hold 96 calutron ALPHA tanks. To minimize magnetic losses and steel consumption, the assembly was curved into an oval 122 feet long, 77 feet wide and 15 feet high.

Note: Groves wanted the first race track in operation by July 1, 1943, and additional tanks to be delivered at a rate of 50 per month so that all 500 would be in production by the end of that year. Roy E. Argersinger, the Berkeley representative for Stone & Webster, insisted that this schedule was impossible. Design of the tanks was not yet complete, and he believed that it would take at least ten days to collect enough information to start scheduling. Groves insisted that the schedule could be met. The general conceptual design of the Y-12 plant was to have five race tracks, each containing 96 tanks housed in three long buildings. The race tracks would be massive, steel, elliptical structures 122 feet long, 77 feet wide, and 15 feet high. Since two tracks would be placed end-to-end on the second floor of each building, the structures would be nearly 450 feet long, of reinforced concrete and masonry. The 48 magnet coils would be connected by a huge bus bar running along the top of the race track and would be energized with direct current from motor generator sets at the ends of the building. Each of the 48 gaps would contain two tanks placed back to back, half facing the outside and half the inside of the tracks. All the internals of the tank—the sources, collectors, and liner—would be fastened to a huge vacuum-tight door which would be installed in the tank by special lifts. More than half the floor area in the building would be occupied by auxiliary and control equipment. Two-story bays on each side of the race track rooms would house the elaborate electrical equipment to supply carefully regulated high-voltage power for the electrodes, liners, and receivers. In these rooms would be located the control panels for each tank, to be manned by hundreds of young women from the surrounding Tennessee countryside. See Figure 5-7, "Control Panels and Operators for Calutrons at Oak Ridge."

The entire ground floor would be occupied by the massive vacuum pumps and by cooling equipment that would service the tanks above. It would also be necessary to build separate chemistry buildings, where the uranium tetrachloride charge material would be prepared from uranium oxide and placed in pyrex or stainless-steel bottles for the sources. Following operation of the tracks, the small amounts of uranium-235 would have to be recovered from each of the collectors by chemical extraction. Most of the charge material would be splattered over the inside of the liner, collectors, and sources and would be recovered by scrubbing and washing in the race track buildings. Facilities in the chemistry buildings would be used to recover the natural uranium from the wash. Thus the plant was conceived in broad outline.

Lack of copper for the large coils to produce the magnetic fields prompted a solution possible only in wartime. Groves drafted 14,700 tons of pure silver from a government vault for the purpose. The silver bars were cast as cylindrical billets that were extruded and rolled into strips by the Phelps Dodge Copper Products Company at Bayway, New Jersey. The strips, $\frac{5}{6}$ inch thick, 3 inches wide, and about 40 feet long, were wound on the magnet coils by Allis-Chalmers in Milwaukee. The huge bus bars of solid silver, roughly a square foot in cross section and running around the top of the race track, were fabricated in Oak Ridge.

Figure 5-7. Control Panels and Operators for Calutrons at Oak Ridge. The Operators, Mostly Women, Worked in Shifts Covering 24 Hours a Day

Late in the summer of 1943 the XAX was ready for testing. After a week of difficulty, it cleared the hurdle for full-scale race track runs.

The first two of five projected race tracks started up in November and failed from contaminated cooling oil; the second was limping in January, but produced 200 grams of uranium enriched to 12% U-235 by the end of February 1944, its fifth of the total goal of one kilogram of enriched uranium per month. By April, four race tracks were functioning, including the repaired number 1. They required constant attention. Many people from the Laboratory helped to modify the units to reach production goals. Responsibility for operation passed entirely to Tennessee-Eastman after the spring of 1944, and the Laboratory staff at Oak Ridge turned their attention to redesigning the calutron system for higher efficiency.

Many at the Laboratory, especially Edward J. Lofgren, thought that a second stage would be necessary to reach the required enrichment. Groves approved the idea. In the spring of 1943, during training at Berkeley for ALPHA operations, design began on the second or BETA stage. Because BETA would have only the enriched product of ALPHA as feed, it would process proportionately less material; its beam therefore did not need to be as broad, nor its dimensions as large, as ALPHA's. BETA design emphasized recovery, not only of the further enriched output but also of the already enriched feed. The first units were tried at Oak Ridge in late February 1944, but the sources had to be redesigned, and even by June difficulties persisted in recovering the precious BETA feed strewn throughout the calutron. Process efficiencies

stayed low; only 4% or 5% of the U-235 in the feed ended up in the output. A better source of enriched uranium feed would have to be found to create the 10 kilograms or so of 90% U-235 that Robert Oppenheimer thought necessary for a bomb.

Note: The calutrons in this second, or BETA stage, as it was called, could be much smaller than those in the first five race tracks, or ALPHA plant. They would be only large enough to process the relatively small amount of material from the first stage. Thus, all the product from ALPHA could be handled by a BETA plant consisting of two magnets, each containing 36 tanks and 72 sources. In most dimensions, the BETA calutrons would be half the size of the ALPHA. The smaller volume of the tanks would make it easier to maintain vacuums, but the shorter running time with each charge would require more rapid pump-down of the vacuum system. The greatest risk in BETA was the possibility of losing the ALPHA product. Since the accumulated product of several ALPHA runs would be needed for one BETA charge, every loss in BETA would amount to a multiple loss in ALPHA.

The "C" shaped ALPHA calutron tank, together with its emitters and collectors on the lower-edge door, was removed in a special "drydock" from the magnet for recovery of uranium-235.

The gaseous diffusion procedure for separation of uranium isotopes, which had consumed more money even than the calutron, had not met its design goals by late 1944. Groves decided that it could not be counted on to produce high enrichment, and that whatever it did produce would have to be supplemented with other slightly enriched uranium and processed through BETA calutrons. To augment the calutron feed, the Manhattan Engineer District (MED) constructed still another plant at Oak Ridge, this one working by thermal diffusion, a method developed by Phillip Abelson.

By 1944 the Allied atomic-bomb program had become a large industrial enterprise devoted to producing plutonium from huge reactors and separating U^{235} isotope from natural uranium.

In the critical production period in the first months of 1945, the calutrons, particularly the six BETAs of 36 tanks each, produced weapons-grade U-235 using feed from the modified ALPHA calutrons, the small output from the gaseous diffusion plant, and whatever the new thermal process had to offer. Virtually all the U-235 sent by courier on the train to Chicago and on to Los Alamos had passed through the BETA calutrons that had raised the isotope concentration to above 80 percent. From these shipments Oppenheimer's physicists assembled the bomb that was to destroy Hiroshima.

Self-Check Questions 5-2

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

- 1. How were the calutrons physically arranged at the Oak Ridge Y-12 plant?
- 2. What were some of the major problems encountered in the Y-12 calutron project?

3. What were the race tracks named and how did they differ from one another?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.3 Identify the uses of the electromagnetic separation process in industry and the required production amounts of enriched uranium.

WORLDWIDE INDUSTRY AND PRODUCTION AMOUNTS

Over the years, calutrons have been regarded—erroneously, some feel—as an anachronistic technology. Essentially a scaled-up version of a mass spectrograph, they are slow and costly to operate. After World War II, they were replaced as the mainstay process for uranium-235 separation by less expensive technologies such as the gaseous diffusion process. Nevertheless, the electromagnetic plant in Oak Ridge during World War II had 1,152 calutrons operated by a now legendary workforce of men and women.

Calutrons are slow and expensive to run. Despite their drawbacks, calutrons can simultaneously produce a variety of high-quality enriched stable isotopes—225 isotopes from 55 chemical elements—that are well-suited for irradiation to produce radioisotopes. Products of these enriched stable isotopes are used for purposes ranging from medicine to materials research to quality inspection. See Figure 5-8.

Figure 5-8. A Technician Adjusts a Calutron Ion Source Used for Stable Isotope Production in United States

United States The Y-12 calutrons produced the first kilogram quantities of highly enriched uranium in 1944. The thermal diffusion plant enriched uranium to 0.86% U-235, which was fed to the calutrons. The thermal diffusion plant was shut down in 1945 after part of K-25 Gaseous Diffusion Plant became operational. A facility designated as S-50 had been constructed to feed partially enriched product to Y-12 to increase the output of the calutron. Because of the lower cost of the gaseous diffusion process, the Y-12 calutrons were shut down in 1946 after all sections of K-25 were operational.

> For uranium enrichment, the production rate of the calutron process was proportional to the feed assay of the ALPHA stage. Thus, if the ALPHA stage assay was increased from the natural assay of 0.711% U-235 to 3% U-235, then the production rate was approximately quadrupled.

On December 13, 1944, a track of 36 calutrons at the Y-12 plant began separating uranium-235 from other uranium isotopes. Another track of 36 calutrons started uranium separations on January 30, 1945. Production of stable isotopes commenced November 11, 1945, with the separation of Copper-63 from Copper-65 in the Building 9731 pilot plant at the Y-12 Plant.

The secrecy of the time, along with an opinion among isotope customers that calutrons were a commercial failure because the gaseous diffusion process was cheaper, put them in disfavor. An "arcane" pricing system also meant that the government isotope sales program lost lots of money. In addition, there was a lack of trained scientists who dealt with the use of isotopes.

Still many researchers hold that, as a producer of stable isotopes for research, industrial, and medical purposes, calutrons are still very useful machines. Calutrons have advantages; they can separate almost any stable isotope, they can separate different isotopes simultaneously, and they may be used to ionize and separate compounds. High-quality enriched stable isotopes produced in calutrons can be irradiated with reactors or accelerators to produce radioisotopes.

In the fall of 1959, Oak Ridge National Laboratory (ORNL) took over operation of the calutrons at Y-12 in Building 9204-3 for its stable isotope production program. Over the years it has operated 30 calutrons for 4 million hours, producing a total of 250 kilograms (550 pounds) of material enriched in 232 different stable isotopes.

ORNL produced Thallium-203 until 1991 when its customers turned to Russia because it was selling the isotope for 10% to 20% less. So the calutrons for isotope production were shut down. However, during the shutdown, the program began selling its reserve supply of Strontium-88 to Amersham International, a pharmaceutical company in England that produces Strontium-89 chloride for relieving cancer-induced bone pain.

Because of their value to research, the Department of Energy (DOE) adopted a National Isotope Strategy in 1994 that promoted production of nonprofitable amounts of rare isotopes.

On December 22, 1994, Trace Sciences International of Ontario, Canada, signed a three-year contract with the DOE to buy stable isotopes in large quantities at a volume discount. These stable, or nonradioactive, isotopes were planned to be distributed to 14 countries for medical, industrial, research, and agricultural uses. In the United States alone, radioisotopes, some of which are produced from stable isotopes, are used for medical diagnosis or treatment of 30,000 to 40,000 patients a day.

ORNL's existing complex of calutrons consists of a rectangular collection of about 30 individual units, located at the Y-12 Plant. Eight units of the calutrons were initially restarted when operations resumed on January 3, 1995, to produce material enriched in Thallium-203. The feed material went to the isotope supplier, which used proton beams in cyclotrons to convert Thallium-203 to Thallium-201. Thallium-201 is a radioisotope used worldwide to diagnose heart disease. The reason for the restart was because DOE had changed its policy, recognizing that some customers that had been buying low-priced stable isotopes from Russia required a

more stable supplier. The commitment by DOE appeared to recapture the stable isotope business that had been surrendered to foreign suppliers, such as the Russians.

Note: The Russians have overproduced in the past, thus lowering prices of stable isotopes and leaving sales revenues alone insufficient to meet operating costs. Congressional hearings during the early 1990s called for annual appropriations to DOE to provide for the production of rare, nonprofitable isotopes for research uses. Annual appropriations were reestablished in FY1995 but were to be directed to other uses, such as the formation of a Mo-99 production facility. Thus, the future supply of rare nonprofitable stable isotopes in the U.S. was once again in jeopardy.

After a successful three-year period of operation supported by sales revenues, the calutrons at ORNL were shut down in 1998 by the DOE because of declining sales. The calutrons provided the U.S. domestic supply of enriched stable isotopes for use in research, medicine, and industry.

Thallium-203, precursor to Thallium-201, is widely used for heart scans. Zinc-68, precursor to Gallium-67, is used for tumor imaging. Strontium-88, a precursor to Strontium-89, is a newly licensed medical radioisotope.

Strontium-89 chloride is the active ingredient in Metastron, a new treatment for cancer-induced bone pain. Strontium-89 is produced for the pharmaceutical company, Amersham International, in European reactors through the neutron irradiation of Strontium-88 targets that were enriched in ORNL's calutrons.

Although ORNL did not certify isotopes for human consumption, their quality assurance documentation and certification of the starting Strontium-88 played an important part in Amersham obtaining Federal Drug Administration (FDA) approval for Metastron.

Another treatment that can relieve the misery of cancer is Rhenium- 188. Tungsten-186 targets enriched in the calutrons are irradiated to produce Tungsten-188, which is then passed through a rhenium "generator" developed in the Health Sciences Research Division's Nuclear Medicine group, led by Russ Knapp. The generator, licensed to Isotope Products Laboratory, Inc., in Burbank, California, produces Rhenium-188, which, when attached to phosphorus compounds, localizes in bone areas. Both Strontium-89 and Rhenium-188 are beta emitters. When they localize near cancerous tumors in bones, the beta radiation reduces tumor cells and fluid accumulation, thus reducing pain-producing pressure on nerve cells running through the bone.

For these specialized and humane purposes, calutrons are a viable source of enriched stable isotopes. Cost is a major reason that isotope supplies have been a concern. For example, prostate cancer patients can be treated with Palladium-103. Palladium-102 must be enriched to have Palladium-103, and that is an expensive process in a calutron. The company TRW had been a producer of palladium targets using the plasma separation process (PSP), which is an ion cyclotron method. TRW discontinued its PSP and ORNL acquired the equipment. In the meantime, available targets of palladium also acquired from TRW were used up.

The biggest barrier to realization of the PSP-calutron combination is the cost of setting it up—several million dollars. Funders for such a project are scarce. Another barrier is foreign competition. Russia possesses some calutrons. Their marketing tactics are very aggressive and their prices are cheap. China also has a few calutrons. But these are hardly dependable domestic supplies. ORNL's calutrons at Y-12 are the only ones in the Western world.

Some researchers say that isotope production may be the most successful "technology transfer" program, so to speak, ever carried out by a national laboratory. In retrospect, it is an astonishing accomplishment that a technology developed more than 50 years ago has changed so little, while the uses of its products—especially in nuclear medicine—are growing by the day.

If supplies of these useful isotopes continue to grow tighter and sources become fewer, ORNL's calutrons may be up and on their way to proving that they are an important national resource—and receive what many scientists feel is some long overdue respect.

Iraq The calutron devices regained some notoriety during the Persian Gulf War when it was determined, largely through the expertise of ORNL veterans such as the late John Googin, that the Iraqis' nuclear weapons program plans involved calutrons for uranium-235 separation. Investigators learned that Operation Desert Storm had derailed an Iraqi nuclear weapons program that was farther along than anyone realized.

> There have been concerns that a small, Chinese-supplied calutron might have been (and might yet be) used for uranium enrichment in Iran. (Spector et al., 1995.)

> Beyond the more-or-less direct connections between radioisotope production and covert weapons programs, there are more general connections. Medical radioisotope production is routinely promoted as one of the most beneficial uses of research reactors. This gives impetus and legitimacy to research reactor programs, despite the potential for covert weapons development. A notable example is Iraq, where the nuclear program was initiated with a small research reactor and a radioisotope laboratory.

Calutrons were built and operated in Iraq in support of Saddam Hussein's fledgling nuclear weapons program. In 1986, Iraq operated its first experimental electromagnetic separator at the Tuwaitha Nuclear Research Center near Baghdad. In 1987, they operated two experimental separators there. Iraq planned to deploy 70 calutrons at one site and 20 at another. In 1990, eight separators were installed and operated at Tarmiya. They were shut down for modifications in 1991. By February 1991, it is believed that air attacks during the Persian Gulf War destroyed Iraq's separator facilities.

Self-Check Questions 5-3

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

- 1. When did the United States produce the first kilogram of highly enriched uranium via the Y-12 calutrons?
- 2. What Tennessee facility supplemented the efforts of the United States calutron process?

3. Why were the Y-12 calutrons shut down in 1946?

4. For uranium enrichment, the production rate of the calutron process was proportional to what?

5. What happened to the Y-12 calutrons after their shut down in 1946?

6. How were calutron devices used in Iraq?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.4 Identify the hazards and safety concerns for the electromagnetic separation process, including major incidents.

HAZARDS AND SAFETY CONCERNS Although it proved effective during the Manhattan Project, the electromagnetic process has several disadvantages that limit its commercial viability. Calutron enrichment is a batch process, which limits its output and requires considerable maintenance. The collectors are removed regularly so the enriched uranium product can be scraped out of them. The calutron tanks and other equipment are periodically washed and cleaned to recover accumulated uranium from their surfaces. Uranium chloride salt oxidizes readily when exposed to air, which creates chemical processing problems in the calutron feed and product material. Table 5-1 identifies some of the hazards associated with the electromagnetic enrichment process.

Table 5-1. Hazards of the Electromagnetic Process

Chemical and Chemical Reaction

The uranium tetrachloride used in calutrons is corrosive, but not as corrosive as the uranium hexafluoride used in other enrichment

that had to be closed. Some of the welds in the magnets gave way and spilled oil on the operating floor. Under the stress imposed by the tremendous magnets, some of the vacuum tanks pulled loose and moved several inches out of line. The failure of rectifier tubes and other electric equipment constantly plagued the early attempts at operation. Even when enough faults could be eliminated to permit some semblance of operation in a few tanks, frequent shorts in the calutrons and clumsy adjustments by inexperienced operators made the attainment of a sustained beam an unusual event.

Under the weight of these failures, the operation of the first ALPHA track came to a halt in the first days of December 1943. The worst problem was the shorts in the magnet coils. When a few of the coils were opened, it was discovered that the shorts were caused not by moisture but by millscale, rust, and other sediment in the cooling oil.

Westinghouse engineers assured Groves that there was nothing wrong with the magnet design. The coils could be expected to operate satisfactorily if the cooling oil could be kept clean. The coils were shipped back to Allis-Chalmers where an attempt was made to clean them without completely dismantling them. Preventative measures were introduced on the installation process; oil piping was carefully cleaned and special oil filters were added to the system.

With the first ALPHA track (ALPHA I) out of operation for weeks, Stone & Webster and Tennessee-Eastman concentrated every effort on ALPHA 2. All oil lines and coils were inspected, oil filters were ordered, and final adjustments were made on calutrons and electrical equipment as soon as they were installed. By January 15, 1944, the first few tanks were evacuated, and the calutrons started. Within a week virtually all the tanks had been operated, although most of them for only a very short time.

Other Problems At first it seemed that routine performance would never be possible with the temperamental equipment. Electrical failures occurred by the hundreds, chemical equipment broke down, and many tanks were turned off for lack of spare parts. Maintenance crews, often on double shifts, were so harried that repair work lagged. An improperly assembled source, a damaged receiver, a cracked insulator, a clogged accelerating slit, a vacuum spoiled by a dead mouse, a corroded chemical tank — thousands of little faults all added up to frustration and disappointment. Only in these production runs was it fully apparent how damaging minor faults would be. The smallest failure inside a tank could require breaking the vacuum seal, a quick repair, and 30 or more hours of pumpdown and bake-out before the operating vacuum was restored.

Self-Check Questions 5-4

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

1. What are some of the disadvantages of the electromagnetic process?

- 2. What chemical hazard was of concern for the electromagnetic process?
- 3. List at least two physical/mechanical hazards for the electromagnetic process.

4. What radiological or criticality hazards were associated with the electromagnetic process?

5. Name at least five mechanical incidents that occurred in the electromagnetic process.

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.5 Describe the principles of the thermal diffusion process.

PRINCIPLE OF THE THERMAL DIFFUSION PROCESS When a mixture is subjected to a temperature gradient, a concentration gradient results. This effect is known as thermal diffusion. If the system is convection-free, the concentration gradient will build until the gain in concentration due to the thermal diffusion effect is exactly balanced by the back diffusion that naturally occurs when a concentration gradient is present. If the system is configured as a countercurrent flow column, an axial multiplication of the simple process enrichment factor results.

> The thermal diffusion separation effect is driven by the thermodynamically irreversible flow of heat from the hot surface to the cold surface. This heat flow may be conductive or radiative. According to the Stefan–Boltzmann law, radiative heat flow is proportional to the difference of the fourth powers of the hot and cold surface temperatures. This fact and the possible thermal instability of the process medium impose practical limits to the temperature of the hot surface.

> The thermal diffusion effect was first observed in liquids by C. Ludwig in 1856. Theoretical work in the kinetic theory of gases in the early part of the 20th century predicted that thermal diffusion would also occur in gases. On theoretical grounds, D. Enskog (in 1911) and S. Chapman (in 1916) each predicted the gas phase thermal diffusion effect before it was observed experimentally by Chapman and F. W. Dootson in 1917. A 1922 paper by R.S. Mulliken first suggested the separation of isotopes by thermal diffusion. In a 1938 paper, K. Clusius and G. Dickel described a new, countercurrent column device in which they made gas phase separations by thermal diffusion. This paper gave the first reports of experimental, gas phase separations of isotopes, namely the isotopes of neon and of chlorine in HCl form. In 1939 these authors reported results from the liquid phase separation of hydrogen isotopes in water as measured in a thermogravitational column.

> The thermal diffusion process as stated above uses a temperature gradient to separate the isotope mixtures. See Figure 5-9, "Schematic Representation of a Thermal Diffusion Column." A temperature difference is created across an annular region between two long, vertical cylindrical pipes. The inner pipe carries hightemperature steam, while the outer is cooled by circulating water. Uranium hexafluoride is kept in liquid form under high pressure in the region between the pipes, and in a very slow process, the lighter

isotope tends to move towards the hot inner wall and then rise to the top of the tube. The thermal gradient causes the light uranium hexafluoride (UF $_{6}$) stream containing more U-235 to diffuse towards the hot wall (inner pipe) and move upward by convection. The heavy $UF₆$ stream containing more U-238 tends to diffuse towards the cold wall (outer water jacket) and move downward by convection. The heavier isotope diffuses outward and is collected at the bottom. This method can produce only very small enrichments, even in tubes as high as 48 feet (14.6 m).

Figure 5-9. Schematic Representation of a Thermal Diffusion Column

The degree of separation obtained depends in part on the magnitude of the temperature gradient across the fluid mixture. Gas phase

mixtures usually have more thermal stability than liquid mixtures. For this reason and to minimize the equilibrium time of the separation system, thermal diffusion is almost always reduced to practice as a gas phase process. The simple process separation factor is usually quite small, so the standard embodiment of the thermal diffusion process is in the form of countercurrent flow columns. Columns can be simply fabricated from cylindrical pipe, which serves as the cold wall, with a central nichrome wire (or calrod heating element) serving as the hot wall. This class of countercurrent flow columns was invented by Clusius and Dickel in 1938. They called the device a "thermogravitational column." Simple devices of the type described by Clusius and Dickel made it possible to consider using thermal diffusion for the separation of isotopes. See Figure 5-10, "Hotwire Thermal Diffusion Column."

In general, thermal diffusion separation factors are extremely small. Simple kinetic theory predictions of the process separation factor for thermal diffusion separation of $UF₆$ in the gas phase gave a value somewhat smaller than that of the gaseous diffusion process. Actual experimental results showed a substantially poorer separation factor than simple theory predicted. Because of this, thermal diffusion separation of uranium isotopes in the gas phase has not been realized.

Figure 5-10. Hotwire Thermal Diffusion Column

Self-Check Questions 5-5

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

- 1. What results when a mixture is subjected to a temperature gradient?
- 2. If the system is convection-free, the concentration gradient will build until what happens?

3. Briefly explain how enrichment of uranium hexafluoride can be obtained through the thermal diffusion process.

4. What affects the degree of separation in thermal diffusion?

^{5.} Between gas phase mixtures and liquid phase mixtures, which one usually has more thermal stability?

- 6. What device did Clusius and Dickel invent to make thermal diffusion for the separation of isotopes possible?
- 7. How do separation factors compare between thermal diffusion and gaseous diffusion?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.6 Describe general facility and component layout of the thermal diffusion enrichment process.

GENERAL FACILITY DESCRIPTION AND COMPONENT LAYOUT Experiments in the early 1940s at Columbia and the University of Minnesota quickly established that the process of using gases was impractical for large-scale separation. Philip Abelson, who worked at the Carnegie Institution, thought of trying liquid rather than gaseous thermal diffusion. Because the facilities were better, Abelson conducted his experiments at the National Bureau of Standards.

> The United States Navy, which by then hoped to concentrate uranium-235 in order to develop a nuclear power plant small enough for submarines, became enthusiastic about the prospects of the method. At the suggestion of Ross Gunn, Abelson transferred in the summer of 1941 to the Naval Research Laboratory in the Anacostia section of Washington, D.C., where higher steam pressures and superior shops were available. At the National Bureau of Standards, the small supply of steam limited Abelson's experiments to columns 12 feet high. In Anacostia, he had a new 20-horsepower, gas-fired boiler which could supply sufficient steam for a 36-foot column.

Procurement problems delayed operation of the first column until November 1941. The column was ruined in an early experiment, and positive results from the operation of the second were not obtained until February 1942. Abelson completed many experiments with six different thirty-six foot columns. He estimated that seven such columns in series would double the enrichment of uranium-235 in the hexafluoride. The disadvantage was that the time required to increase the enrichment even as high as 50 percent would be impractically long. He then experimented with two 48-foot columns operating in series. About two kilograms of hexafluoride per day were processed.

Note: Before the end of 1941, Gunn and Abelson had placed a small order with the Harshaw Chemical Company in Cleveland, Ohio, to acquaint it with the process. The Eger V. Murphree's Planning Board supplemented this early in 1942 with a contract to build and operate a pilot plant producing 10 pounds of hexafluoride per day. By spring, the Harshaw plant was operating, and Du Pont was also experimenting with the process.

> It should also be noted that early plants had no moving parts and no valves in the hexafluoride system. The process liquid

moved by natural convection. The temperature and pressure conditions of the hexafluoride made it possible to stop its flow simply by freezing the supply pipe with a handful of dry ice. A rough calculation showed that to construct a plant producing one kilogram of fully enriched uranium-235 per day would require 18 months and \$75 million.

In November 1943, Abelson received Navy authorization to build a 300-column plant in Philadelphia, Pennsylvania. Construction started in January 1944 on a 100 column plant, which was to be operated as a seven-stage cascade before the larger plant was built.

On June 27, 1944, H. K. Ferguson Company management of Cleveland, Ohio, signed a War Department contract to construct a thermal diffusion plant within 90 days at the K-25 plant site located in Oak Ridge, Tennessee. This plant was designated as the S-50 plant. The day before, Admiral Ernest J. King had ordered blueprints of the Philadelphia pilot plant sent to the contractor. After studying these drawings and examining Abelson's 100 tube plant at the Navy Yard, the Ferguson engineers concluded that they could do little more than build 21 exact duplicates of the Philadelphia plant and tie them together with steam lines running to the K-25 powerhouse.

The one critical procurement item was the 48-foot columns in which the thermal diffusion process occurred. It was one thing to make 100 columns in a Navy shop and something else to fabricate more than 2,000 of them commercially. Extremely uniform tubing was required to maintain the critical annular spacing between the inner nickel and outer copper tubes. Perfect roundness and a tolerance of 0.002 inch was difficult to attain especially when copper and nickel tubing could not be drawn in 48-foot lengths. Only after canvassing 21 manufacturers did Ferguson find two contractors willing to accept the assignment. The Mehring & Hanson Company of Washington, D.C., and the Grinnell Company of Providence, Rhode Island, quickly devised methods of welding and soldering shorter lengths of nickel and copper tubing. They also learned how to maintain the critical annular spacing between the tubes by welding small nickel buttons to the inner tube to act as spacers. Fastening the tubes together was a tricky process requiring the use of hot nickel and cold copper to reduce thermal stresses during operation. Using trial and error methods, the fabricators were soon able to reduce the number of failures and increase production in each plant to 50 columns per day. A huge barn-like structure of black-coated siding more than 500 feet long, 80 feet wide, and 75 feet high was erected. See Figures 5-11 through 5-13 for a visual representation of the S-50 plant site, its thermal diffusion columns, and their components.

Figure 5-11. S-50 Plant

Figure 5-12. Thermal Diffusion Column

Figure 5-13. Liquid Thermal Diffusion Column

The S-50 plant was constructed to provide low enriched feed (0.86% to 0.89% U-235) for the Y-12 electromagnetic spectrometers (calutrons). In August 1945, less than a month after the bombing of Hiroshima, the thermal diffusion plant in Oak Ridge was shut down. A lavish amount of money was spent for a marginal through-put for the thermal diffusion process.

Self-Check Questions 5-6

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

1. What were some of the problems incurred in the building of thermal diffusion facilities in the 1940s?

- 2. When using two 48 foot columns, how many kilograms of hexafluoride per day were processed?
- 3. True or False. Early plants had no moving parts and no valves in the hexafluoride system.
- 4. Why was the S-50 thermal diffusion plant built in Oak Ridge, Tennessee?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.7 Identify the uses of the thermal diffusion process in industry and the required production amounts of enriched uranium.

INDUSTRIAL USE Although a large-scale, liquid phase thermal diffusion process uranium enrichment plant was deployed in the Manhattan Project, the process was never again used for uranium isotope separation. The main reasons are the long plant equilibrium time, if liquid $UF₆$ is used as the process fluid, and the fact of its extremely low thermodynamic process efficiency. It has been said that if the enriched product from a thermal diffusion uranium enrichment plant were used as fuel in a civilian power reactor, that reactor could not supply enough energy to power the enrichment plant producing its fuel.

> In the Manhattan Project, the liquid phase thermal diffusion plant was designated "S-50." It operated for about a year before being shut down, decontaminated, and dismantled. During its operation it enriched normal assay UF₆ to about 0.85 weight percent ²³⁵U. The resulting product was used as feed for the electromagnetic separators at the Y-12 plant. It was built because a 300-megawatt (MW) generating station, built to support the gaseous diffusion enrichment plant, was able to raise steam long before the gaseous diffusion plant could utilize its electric power. Work done at the Naval Research Laboratory in Washington, D.C., on liquid thermal diffusion of $UF₆$ showed promise and the water-cooled, steam-heated columns could be built quickly. The decision was therefore made to build a thermal diffusion enrichment facility for pre-enriching the feed for the Y-12 electromagnetic enrichment plant. Enrichment to 1% in 235 U was the goal. If that could have been accomplished, it would have increased the Y-12 output by 40% because there is 40% more 235 U per kilogram of feed at 1% enriched than in natural assay feed.

> Isotope separation using gas phase thermal diffusion has been widely practiced in the United States and abroad. U.S. applications have included separation of both stable and radioactive isotopes. These applications were uniformly restricted to very low production amounts (gram per day amounts). Thermal diffusion is ideally suited to this scale of production because its process equipment is compact and very simple and its energy costs are neither large nor dominant in process economics. For this application, thermogravitational columns a couple of inches in diameter and 20 feet or so long can be used. The columns can be air-cooled and mounted vertically on wall racks. Intercolumn flow occurs by thermal siphon and no operator attention is required.

The magnitude of the thermal diffusion effect in gases depends sensitively on the intermolecular force potential assumed in deriving the gas kinetic equations. A simple and often-used intermolecular force model for gases assumes point masses and inverse power-law repulsion between molecules. For this force model, the thermal diffusion effect causes the light component to concentrate at the cold wall if the exponent is less than five. For spherical molecules, if the power law exponent is exactly five, the thermal diffusion effect vanishes identically. For this reason, gas kineticists have used thermal diffusion as a means of elucidating the intermolecular force potential between molecules of many types. There is a substantial body of literature reflecting these studies.

Self-Check Questions 5-7

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

1. Why is thermal diffusion no longer used as a uranium enrichment process?

2. What enrichment amount did the S-50 plant in Oak Ridge produce and how was it used?

3. How has gas phase thermal diffusion been used in the past?

You have completed this section. Please check off your progress on the tracking form. Go to the next section.

Learning Objective

When you finish this section, you will be able to:

5.1.8 Identify the hazards and safety concerns for the thermal diffusion process, including major incidents.

HAZARDS AND SAFETY CONCERNS

- **Chemical Hazards** Thermal diffusion uses large quantities of liquid UF_6 . UF₆ is chemically reactive. It reacts vigorously with most organic compounds at a rate that is temperature dependent. Common reaction products include UF_4 , HF and carbon. It reacts with most metals to form a fluoride of the metal plus a lower uranium fluoride such as UF_4 or UF_5 . In the case of a few metals (nickel is an example), the fluoride forms a tightly adhering coating that protects the metal from further reaction. In the presence of moisture $UF₆$ is immediately hydrolyzed, forming the solid uranyl fluoride $(UD₂F₂)$, and hydrogen fluoride (HF). Hydrogen fluoride gas is very toxic, but perhaps the major process safety issue with this reaction is that UO_2F_2 is less dense than UF_6 and hence occupies more space than the UF₆ that produced it. If this reaction occurs in a confined space, process equipment can rupture hydrostatically. This problem is particularly acute when the UF $_6$ is present in solid form where it is essentially incompressible. Water cooling of process equipment must therefore be carefully engineered to ensure isolation of the cooling water from the UF₆ process fluid. Liquid UF₆ is pressurized and any confinement leak rapidly flashes quickly engulfing the area in a white cloud.
- **Mechanical/Physical Hazards** Mechanical/physical hazards for the thermal diffusion process mainly relates to the use of steam. In earlier plants, hissing sounds of escaping high-pressure steam drowned out all conversation and enveloped the column racks. Also, the fabricator had to develop a new type of connector between steam lines and the columns to eliminate excessive leakage.

Incident On September 2, 1944, at the S-50 plant in Oak Ridge, the mechanical failure of a tank containing uranium hexafluoride killed two operators and injured several others. It did not take Phillip Abelson long to make repairs, but by that time the first rack was nearing completion in the S-50 plant.

Self-Check Questions 5-8

Complete the following questions. Answers are located in the answer key section of the Trainee Guide.

1. What happens to UF₆ in the presence of moisture?

2. What problems can UO_2F_2 and HF cause in the UF₆ process system?

- 3. What is a main mechanical/physical hazard related to the thermal diffusion process?
- 4. Briefly describe the results of the incident that occurred at the S-50 plant in Oak Ridge, Tennessee, on September 2, 1944.

It's time to schedule a progress meeting with your administrator. Review the progress meeting form on the next page. In Part III, As a Regulator, write your specific questions to discuss with the administrator.

Progress Review Meeting Form

Date

Scheduled: Constanting Cons

I. The following suggested items should be discussed with the administrator as to how they pertain to your current position:

- Principle of the electromagnetic separation module
- History of electromagnetic separation research and development
- \cdot Industrial use
- Hazards and safety concerns
- Principle of the thermal diffusion module
- History of thermal diffusion research and development
- Hotwire thermal diffusion column
- Liquid thermal diffusion column
- \cdot S-50 plant
- \cdot Industrial use
- Hazards and safety concerns

II. Use the space below to take notes during your meeting.

III. As a Regulator:

- What should I become most familiar with in the electromagnetic separation process?
- Is there any documentation that I should read regarding electromagnetic separation?
- How does electromagnetic separation compare to other enrichment processes?
- What should I become most familiar with in the thermal diffusion process?
- Is there any documentation that I should read regarding thermal diffusion?
- How does thermal diffusion compare to other enrichment processes?

Use the space below to write your specific questions.

IV. Further assignments? If yes, please note and complete. If no, initial completion of progress meeting on tracking form.

Ensure that you and your administrator have dated and initialed your progress on your tracking form for this module. Go to the module summary.

Congratulations! You have completed the fifth module of the Uranium Enrichment Processes Directed Self-Study Course. Go to the Self-Study Course Process in the Trainee Guide. Ensure completion of steps H and I.