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Electromagnetic Separation of Isotopes at Oak Ridge

An informal account of history, techniques, and accomplishments.

L. O. Love

For almost 30 years the Atomic Energy Commission has been separating the isotopes of the elements and supplying these unusual materials at cost to research workers in all fields all over the world. No great publicity has attended this work and by now it is almost taken for granted; nevertheless, it has a history with associated anecdotes, and in this article I shall attempt to outline the growth of the effort and describe the present system as it appears to one who has been close to the operational level.

Background

In 1943, the Manhattan Engineering District entered into a contract with the Tennessee Eastman Corporation to operate a large and secret electromagnetic facility known as Y-12 near the newly created town of Oak Ridge, Tennessee. The purpose was to obtain large quantities of uranium-235 free of the more abundant isotope uranium-238. The process was one of mass spectrometry and it was one of the great wartime efforts of the Manhattan District. By 1945, the electromagnetic isotope separations program was functioning at its peak with nearly 25,000 people and more than 1100 separating units. When the plant was in its early stages and, in one layman's terms, "Ph.D.'s were as common as three-cent stamps," a practical-minded East Tennessean commented that he didn't know what was being made here, but he knew one thing—it could be bought cheaper somewhere else. Indeed, this was to be the case, because by mid-1945 the gaseous diffusion process had demonstrated that there was a cheaper way of obtaining ^{235}U and soon after the cessation of hostilities the electromagnetic plant was declared obsolete and the shutdown of facilities was initiated.

During this period representatives of the Manhattan Engineering District and Tennessee Eastman Corporation met to discuss the possibility of using a part of the plant to supply usable quantities of enriched isotopic materials for basic and applied research. Soon afterward there was an exchange of correspondence between E. P. Wigner of the Clinton Laboratories (now the Oak Ridge National Laboratory) and A. V. Peterson of the Manhattan Engineering District.

Wigner wrote, ". . . to review some of the things which were brought out in our conference with you and Drs. McDaniel and Aebersold. . . . In our opinion the work now being done . . . at the Y-12 Plant is and promises to continue to be scientifically one of the most important projects now under way in this country. We should have, as the very basis of future work in nuclear physics and chemistry, knowledge of the various cross-sections of pure stable isotopes. Eventually separated isotopes of the elements may provide invaluable raw material for the production by pile or other irradiation of radioisotopes of value in science, medicine and industry. Since we believe that the stable isotope program at Y-12 is today scientifically more important and soon will be more important on every count than the uranium isotope separation, we wish that greater emphasis could be placed on it."

Peterson replied, "As you no doubt are aware the District has held discussions, in line with the comments made in your letter, and . . . you will be glad to know that Dr. Clarence Larson has been placed in charge of the overall stable isotopes program at Y-12. Both he and Dr. Keim, who supervises the electromagnetic phase, are enthusiastic about the program and will promote it as fully as possible."

These letters were a result of discussions in late 1945 and early 1946 between members of the Electromagnetic Process Improvement staff of the laboratory and P. W. McDaniel, who was largely responsible for initiating the stable isotope separation effort. Following this exchange, a pilot plant, which contained four separators or "calutrons" (*1*), was devoted to processing the stable elements with multiple nuclidic species.

When the shutdown operation was finished, only two of the nine Y-12 buildings housing calutrons were retained intact—the pilot plant with two beta and two alpha separators (*1*) and a production building containing 72 beta separators. The immediate plan was for the pilot plant to be used for the separation of stable isotopes, while the production building would be used for improving the process and for mak-

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ing special uranium separations such as the enrichment of ^{234}U.

Since then, some 200 kilograms of highly enriched isotopes embracing more than 250 stable nuclidic species, plus a number of radioisotopes including those of beryllium, bismuth, calcium, thorium, uranium, plutonium, americium, curium, nickel, and samarium, have been made available from the facility, which is now part of the Oak Ridge National Laboratory. The separated isotopes provide the basis for worldwide investigations in nuclear physics, chemistry, the earth sciences, biology, and medicine. Isotopic purities range from a few times natural abundance to a purity level in which unwanted isotopes are present in no more than tenths of a part per billion.

The stable isotope program began in an atmosphere of turmoil when the Y-12 plant was being dismantled and employment drastically cut. To those of us involved it was apparent that costly equipment items soon would disappear, and it was equally apparent that some of the existing equipment would be needed in the future if we were to process 250 different isotopes from over 50 elements. Action was required without time for formalities, and as matters evolved the responsibility seemed to fall on me. After Tennessee Eastman left Oak Ridge, the operation of Y-12 was assumed by the Union Carbide Corporation, and I think that the only time my name was ever mentioned in their New York office was when I was offered as an example of people who were making it difficult to dismantle Y-12. Others would take the equipment to the salvage yard; I would bring it back.

In retrospect, it is clear that my procurement methods did present problems to those implementing the nation's salvage policy. For our purpose, used equipment and surplus in stock had to be procured on the basis of projected need rather than current usage. Funds were not available for stockpiling these materials and regulations would not permit the use of current funds to supplement subsequent years' operations. Consequently, the only solution was to obtain the material on an "informal" basis. Presumably, this could have been done only by people who made their own interpretation of the broad concept of national policy relating to the reduction of wartime inventories and who, fortuitously, were blessed with a sympathetic management.

For example, at one time we were instructed to submit specifications for a reorder of ionization gauge tubes because the supply was exhausted, but we knew that several thousand surplus tubes were stored in a warehouse that also housed some of our equipment, with only a wire fence separating the two storage areas. Our reasoning was elementary—if the tubes were recorded as lost, they could be slipped across the fence to our side of the warehouse. Some months later the transfer was discovered by means of an efficient storehouse record system, and we were instructed to return the ion gauge tubes. However, we still had the form requesting specifications for a new order and, with a little negotiation, we were allowed to retain a good supply of the surplus ion gauge tubes, which actually kept us supplied until 1964. Although our approach was naive and our methods sometimes crude, 25 years later we still have a stockpile of basic equipment that enables us to operate up to 40 separators, changing from one element to another with ease.

The electromagnetic process is undoubtedly the most versatile known means of separating isotopes. Development of the calutron started in 1941 under E. O. Lawrence at the Radiation Laboratory, University of California. Use was made of the magnetic field of the 37-inch cyclotron to disperse the ^{235}U and ^{238}U after a deflection through 180°; this increased the purity of ^{235}U by a large factor and proved that one could contain useful qualities. This method of separating isotopes was not new at the time; indeed its principles go back to the beginning of the century. The 180° deflection method was applied to mass spectrometry by Dempster in 1918 (*2*), using a focusing principle recognized by Classen in 1908 (*3*). The focusing is of course important for intensity. Semicircular deflection was also used by Nier *et al.* (*4*) in their historic first separation of the uranium isotopes in 1940 by using a separator with a 7-inch radius. The magnitude of the calutron development is evident when one considers that the total quantity of ^{235}U separated by Nier and his co-workers was about 10^{-8} gram, whereas before the Y-12 electromagnetic plant was shut down, kilogram quantities had been produced.

Since the inception of the stable isotope program some of the multinuclidic elements have been processed only once; other separations have been repeated as many as 19 times. Impurities consisting of isotopes separated by one mass unit have, on special occasions, been confined to less than 1 part per million and, in some cases, to the parts-per-billion range. Enrichment factors for various isotopes range from about 30 to as high as 80,000 in a single pass (*5*).

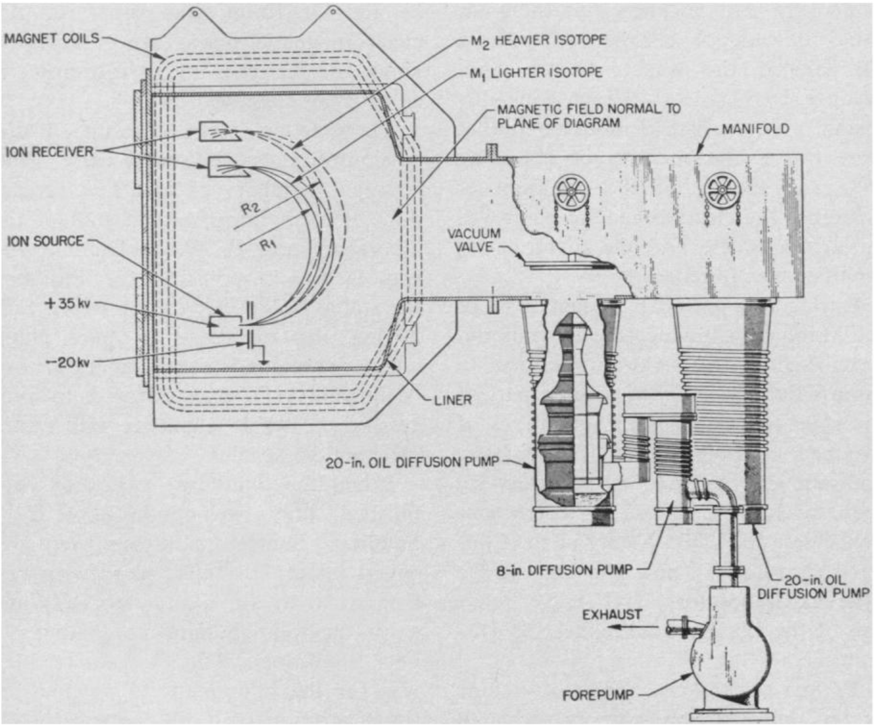


Fig. 1. Diagram of a beta calutron separator. The ion source, accelerating system, receiver, and semicircular focusing ion paths are sketched in.

Elementary Theory and Basic Equipment

As a particle of charge e and mass m travels across a magnetic induction of strength B, it moves in a circular path, and the centrifugal force can be equated to the magnetic deflecting force

$$\frac{mv^2}{r} = Bev$$

where v is the velocity of the particle and r is the radius of its path. If the particle is accelerated from rest in an ion source by falling through a potential difference V, then $Ve = \frac{1}{2}mv^2$. Eliminating v between the two equations, one obtains

$$r = \frac{1}{B}\left(\frac{2mV}{e}\right)^{1/2}$$

so for particular values of B, e, and V, the radius of the orbit varies as the square root of the mass of the particle. Thus, the ions of the isotopes of an element can be separated, the separation being maximized at the 180° deflection point (see the orbits sketched into Fig. 1). Taking differentials, one sees that $2\Delta r/r = \Delta m/m$, so for the separation of uranium in the alpha calutron ($r = 123$ centimeters, $\Delta m = 3$, $m = 238$) the 235 and 238 beams are separated by $2\Delta r = 1.54$ cm.

The Oak Ridge electromagnetic plant used these classic principles on a grand scale. A separator tank for the alpha-size calutron measures 12 feet by 8 feet by 28 inches and weighs 10 tons. Only two of these now remain, and are located in our pilot plant building (Fig. 2). The beta tanks are 6.5 feet by 5 feet by 23 inches and weigh about 9 tons. The magnetic field strength for uranium separation is 3200 gauss in the alpha separators and 6400 gauss in the betas. The weights of the conducting windings are in the range 10 to 17 tons for both alpha and beta coils.

The basic components of a calutron ion source are a box, a slit, and a hot filament. The box is made of graphite and measures about 1⅛ by 1 by 6 inches, and the filament (a hairpin of tantalum 11/64 inch in diameter) is set at one end. The whole is arranged so that the electron stream from the filament runs the length of the box, in a direction parallel to the calutron's magnetic induction. The slit is usually about 3/16 inch wide, and is in the side of the box, parallel with the electron stream. When volatile feed material is admitted to the box, ions are formed in the electron stream and are extracted through the slit (across the magnetic field) by the accelerating electrodes indicated in Fig. 1, just outside the slit. The acceleration system employs both acceleration and deceleration for positive ions, as indicated by the applied voltages; this maximizes the ion extraction, improves focus (partly by modifying the shape of the plasma surface from which the ions are drawn), and provides a final ion energy suitable for deflection. The ion beam currents are typically tens of milliamperes. A modern ion source comprises many developments beyond the basic concepts just mentioned and an idea of what one looks like may be obtained from Fig. 7, although this is a small, specialized source.

The charge materials selected for the ion sources must meet requirements of chemical purity, freedom from moisture, appropriate vapor pressure, and stability in a vacuum, and they must be dense enough to stay in place during pumpdown. The elemental forms are used when possible, but since this is not generally the case we seek simple compounds that have a vapor pressure of about 50 micrometers at some convenient temperature, and thus we can control the feed by controlling the oven temperature. A good trick is to use the oxide of the element with the addition of carbon tetrachloride (*6*). This allows one to regulate the separation merely by turning the CCl_4 valve.

The receivers for the ions have to be tailored to match the element under separation. One is shown sectioned in Fig. 3. A well-focused ion beam of 40 thousand electron volts (40 kev) and only a few milliamperes striking a collector causes severe erosion and results in wear of the collector pockets and loss of valuable separated isotopes. Because of this sputtering problem, the pockets are designed with consideration given to the chemical effect of the beam on the pocket material as well as properties like the vapor pressure of the element being processed and the chemical problems associated with recovering the enriched isotope. Usually carbon, copper, or aluminum is used as structural material, with carbon being the preferred element since it is easy to machine into complex shapes and because it generally simplifies chemical recovery. The metallic collectors must be used for volatile elements such as selenium, sulfur, rubidium, chlorine, and bromine because of the cooling problem.

The focus of the different isotopic ion beams is one of the more delicate tasks associated with the operation of the calutron and is dependent on electric as well as magnetic parameters. One factor early recognized was the necessity for space-charge neutralization of the ion beam to avoid blowup because

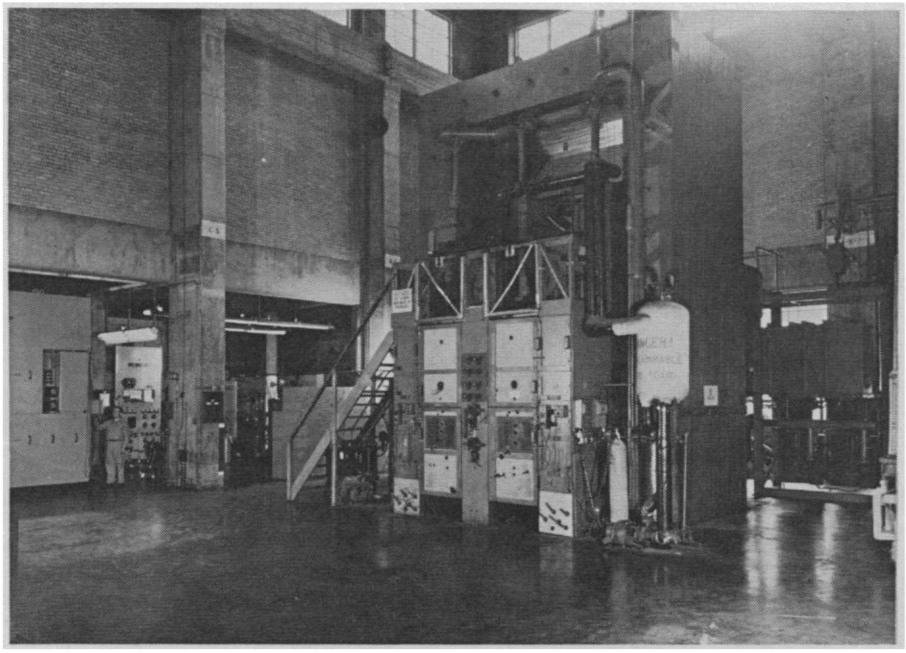


Fig. 2. The alpha calutrons in the pilot plant building. The high structure is the magnet yoke; the vertical pipes lead cooling oil to the magnet coils. The paneling in front of the separators proper serves as high-voltage protection. Vacuum pumps are behind the structure (right), and a "cubicle" supply for the accelerating power is at the extreme left.

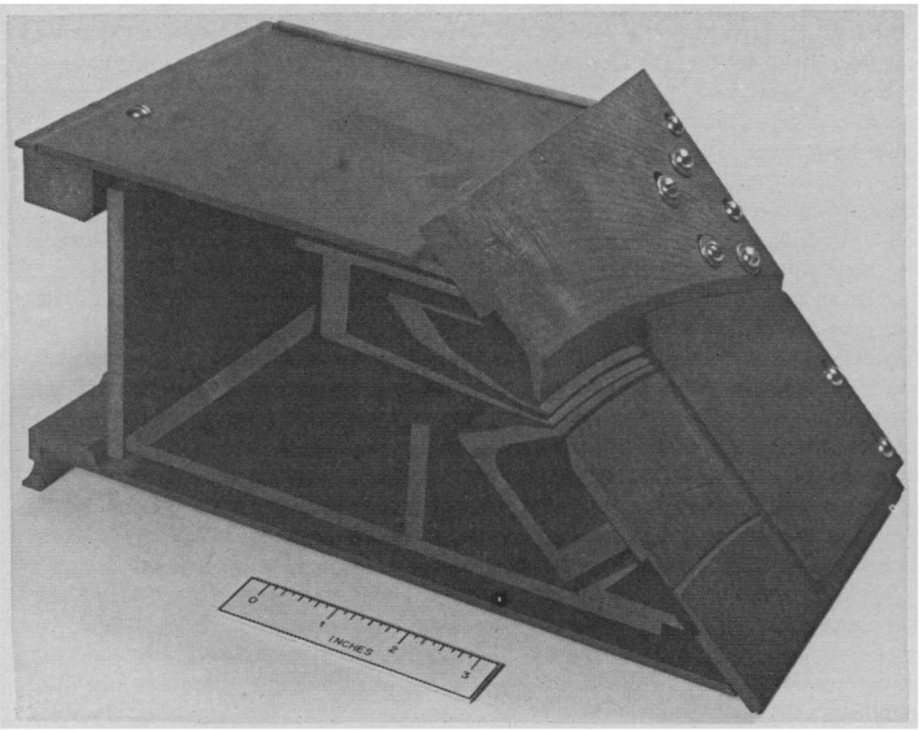


Fig. 3. Receiver designed for plutonium, sectioned to show the pockets. The bottom slit (right) is for monitoring; the four grouped slits receive the isotopes 239, 240, 241, and 242 from bottom to top. The material is graphite.

of electrostatic repulsion. An answer was to introduce a population of electrons coming from ionization by the beam of residual gas in the vacuum tank. The degree of neutralization thus obtained is adequate if all adjustments can be made precisely enough; however, if space charge cannot be controlled, the beam will exhibit a random and rapid fluctuation known as "hash" that is now thought to be caused by beam-plasma instability. This condition can be rectified by increasing the pressure in the beam region—an action which increases losses because of particle scattering and neutralization. Thus, ion formation, beam transmission, and focus are accomplished by a series of compromises, but once good beam conditions are achieved and the beam is going into the collector pocket, only occasional adjustments are needed to keep the operation steady.

Early Separations in the Pilot Plant Building

One of the early problems with the calutron processing of uranium was that the vapor of any element finding its way into the ion source was ionized, and thus unwanted ions made up a part of the beam spectrum. Stray beams of ion source components such as iron, chromium, nickel, and copper not only limited the uranium output from the ion source but literally cut the operating equipment to pieces by their sputtering action. The intensity of these unwanted beams prompted remarks that the separators would be fine for elements other than uranium, and late in 1945 the two beta calutrons in the pilot plant were used to separate the isotopes (63 and 65) of copper—our first stable isotope collection.

The first publication (*7*) based on the use of calutron-separated isotopes states: "The availability of enriched copper isotopes in the Manhattan Project has now made possible a positive assignment of the 2.6 h nickel isotope to a mass number of 65." This first investigation provided tangible evidence that materials from the calutron were unique for physical investigations, and such people as W. A. Arnold, R. N. Goslin, C. P. Keim, C. E. Larson, and P. W. McDaniel should be remembered for their efforts to save a portion of the electromagnetic plant which enabled the nation to provide separated isotopes for programs in the fields of biological, medical, and physical research.

The accuracy of the foresight of these men and others may be exemplified by the extent of present interest by the medical professions in such isotopes as molybdenum-98 and mercury-196. These enriched isotopes, when exposed to neutrons in nuclear reactors, produce radioactive technetium-99m (m = metastable) and mercury-197, which have become valuable in the early diagnosis of malignant tumors (especially brain tumors) through recently developed scanning techniques in nuclear medicine. Sales of these two stable isotopes alone exceeded $300,000 in each of fiscal years 1971 and 1972.

When I came to Oak Ridge in 1943, operation of the calutron had just begun. During my years in isotope separation, many interesting and amusing incidents have happened. One is related to the collection of mercury, where the interest was in providing milligram quantities of 98 percent mercury-202 for the late W. F. Meggers of the National Bureau of Standards; the isotope was to be used as the basis for an optical wavelength standard. We had developed a technique of retaining the mercury by allowing the mercury ion beam to strike a silver collector pocket, so that the mercury would be held by amalgamation. The trouble was that the silver did not show sufficient preference for the separated mercury ions over the normal mercury vapor in the low-pressure calutron tank atmosphere. We were able to get the ^{202}Hg enriched to about 95 percent, but further efforts to improve the isotopic purity were unrewarding. The simplest way to reduce the neutral mercury vapor background was to refrigerate the calutron liner. A cooling system was designed to use Dry Ice, and a search started to find a compound with appropriate specific heat and viscosity for use as a transfer medium. The compound ethanol met the specifications, and an order for 35 gallons was placed. We were operating on a continuous basis in those days, without regard to holidays or weekends, and this happened to be the Christmas season. Shortly after the order was placed I began to receive calls inquiring about the uniqueness of an experiment that required the properties of this particular substance. I was so involved in trying to lick the technical problem that the coincidence of the request for ethyl alcohol and the festive season did not occur to me. The stockroom attendant did see the coincidence, particularly in dry Tennessee, and showed his acumen by billing the entire 35 gallons in pints. To top it off, the operators had been instructed to be extra safe in venting the system to avoid a pressure buildup. It has always been difficult to convince listeners that the alcohol ordered a week later on New Year's Day was to resupply the cooling system because of excessive evaporation. The magnitude of the refrigeration effort is better appreciated when it is understood that 288,000 pounds of Dry Ice were used to get the 98.3 percent ^{202}Hg that was needed. Today we would make the same collection by allowing the mercury ion beam to strike a thin film of oil; the ions continuously carbonize the oil, the carbon retains the isotope, and the thin layer of oil protects it from the normal mercury vapor (a process which was developed by W. A. Bell and A. M. Veach). For a similar collection the oil would cost $1 and the isotopic purity of the 0.15 percent abundant ^{196}Hg would be, for example, increased from about 5 percent to 50 percent.

By using the four regular calutrons in the pilot plant and extending the temperature range of the ion sources from the uranium range of 350° to 450°C downward to room temperature and upward to 2800°C, all elements with naturally occurring stable isotopes had been processed at least once by the mid-1950's, except for osmium which was done in April 1960.

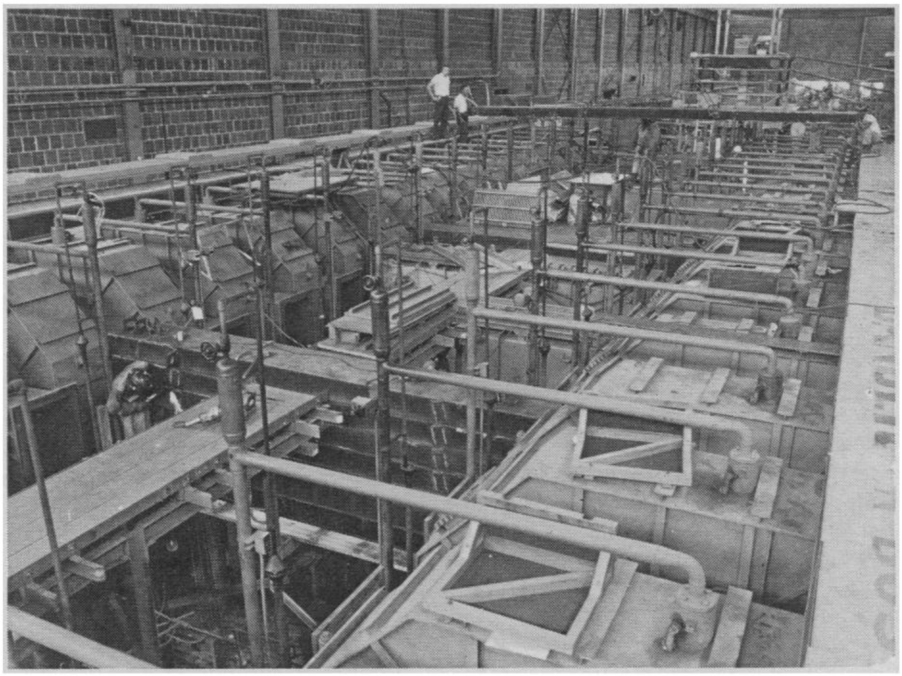


Fig. 4. Segmentation of one of the beta "racetracks" in 1959. In the background, a crane lowers a segment of one of the new cross yokes. A completed cross yoke is shown in the foreground. The boxlike structures contain the magnet coils, and the open vacuum manifolds of three separator tanks can be seen left of center.

Expansion of Capabilities; Design Improvements

In 1959, the need for enriched stable isotopes had increased so much that the quantities required could not be supplied by the four pilot-plant calutrons, and the Atomic Energy Commission decided to devote the 72 beta calutrons in the production building to the general isotope separation program. These separators are positioned in two rectangular tracks, each track containing 36 calutrons in two 100-foot-long parallel arrays. The two sides of each track are joined across their ends by 30-foot-long, 160-ton iron yokes as required to make an overall magnetic circuit closed except for the gaps that hold the calutrons (Fig. 4).

To separate the isotopes of a particular element, one would seldom need all 36 calutrons in a track; rather we wished to be able to devote several calutrons to one element, several others to another element, and so on. This would require different magnetic field strengths in the different sections of a track, but the monolithic nature of each track, powered by a single motor-generator, was an obstacle. To gain the desired flexibility we subdivided the tracks into four segments with three iron cross yokes weighing 80 to 100 tons each (Fig. 4), a separator being sacrificed at each end of each cross yoke.

Additional motor-generators for exciting these newly formed sections came from various places, and all were in used condition. One generator previously supplied power for the elevators in the city of Los Angeles, another came from a sugar refinery, and a third was initially used to form wooden components in the body plant of the Ford Motor Company. In all, we now have ten generators with an installed direct-current capacity of 33,000 amperes at 350 volts.

Throughout the track modification we cut corners wherever we could, using our own engineering experience and intuition for quality assurance in such matters as acquiring the necessary motor-generator sets. When all was ready, we faced a severe test. Two elements held top priority: calcium and lead. Calcium-46 was needed to make ^{47}Ca for use in an international program on the early identification of bone malig-

Fig. 5. Dismantling the silver alpha coils for return to the Treasury Department, 1969.

nancies, and ^{204}Pb was required for an age-of-the-earth experiment. One element requires a high field and the other a low field, but we processed them simultaneously in adjacent track segments quite successfully, showing that the magnetic circuits in the two segments did not interfere and that our model magnet tests had been sound.

Looking back at the track modification, we put 24 separators into operation instead of the budgeted 16, we installed three cross yokes instead of the two that had been planned, we bought a used but good motor-generator from the city of Los Angeles, we built a chemistry laboratory for processing the isotopes—and we returned $54,000 to the Atomic Energy Commission.

Those Silver Coils

Originally all of the alpha coils (about 800) and about half of the beta coils (about 150) were wound with silver, a total of about 15,000 tons of silver being used in the electromagnetic plant. When the U.S. Treasury Department was approached about borrowing hundreds of tons of silver for the magnet windings, their spokesman replied, "Sir, we measure our silver in ounces, not tons." In keeping with this philosophy, when the bus bar connections were being made, a guard was always present to pick up the filings.

After the war, the heavier silver bus bars and most of the silver in the coils were returned to the Treasury Department when the plant was being dismantled. Some of the silver coils, however, were retained for several years. Each year, the Treasury Department politely requested its silver back, the local AEC operations office conferred with the laboratory, and the request was politely declined on the grounds that (i) the laboratory saw a finite chance that the coils might be economically reused, unique as they were; (ii) the silver was safe, being massively enclosed in welded oil-cooling jackets and kept in a guarded area; and (iii) it would cost a lot to ship it back. This routine went on until 1968. By that time the Thermonuclear Division at Oak Ridge had emerged as the sole conceivable future user of the coils (for plasma work in controlled fusion), and as that work developed in other directions, A. H. Snell (then assistant laboratory director and director of the Thermonuclear Division) decided that the silver should be released. The coils were then cut up and returned to the Treasury Department (Fig. 5), with no publicity until the operation was complete.

Fortunately, most of the beta coils had been made of copper and replacing the three beta coils in the pilot plant was only a matter of exchanging them. However, we had almost 37 tons of silver in the three alpha coils, and since there were no alpha coils with copper windings we had to fabricate them. The existing coils were dismantled and rewound with copper conductor taken from beta coils that had been saved when the plant was dismantled. A full alpha coil winding contains over a mile of copper conductor and weighs more than 16 tons.

Technological Improvements; Some Special Separations

The longer we have been associated with the electromagnetic method of isotope separation, the more we realize how fortunate it was for the war effort that uranium was the element requiring separation. The vaporizing temperature of the tetrachloride, the separation of three mass units between isotopes, the ionization potential, the lowered work function of a uranium-coated tantalum filament—all of these favorable aspects and more make uranium separation a pleasure compared to most other separations. As examples of specific problem elements one can cite barium, strontium, and cerium for excessive ion-source sparking; mercury for its vapor pressure problems; iron, nickel, copper, zirconium, and hafnium, which deposit in the arc region and block off all ionizing electrons; the gases for their inherent collector problems; and the heavier elements with their small one-mass-unit separation between adjacent beams. Each element is almost unique, requiring specific operational parameters and techniques if not specific equipment. In general, characteristics that are desirable at the ion source are intolerable at the collector: one wants reasonably high volatility in an ion source feed, but no volatility of the collected species. Thus, an iron halide is used as feed and the isotopes are collected as elemental iron. Fine, but what about the reactivity of finely divided iron isotopes with residual halogen, and subsequent volatilization? With iron the problem is not too bad; with titanium, vanadium, and similar elements we found it intolerable for low-abundance isotopes until the advent of calcium pumping (see next paragraph). In like vein, for some 20 years no suitable charges were known for the elements of the platinum group. Separation was achieved only by vaporizing the elements themselves into the arc column at temperatures up to 2800°C. Since electron bombardment was used as a heating method, the charge container, which was made of graphite, often lasted only 8 to 10 hours. Needless to say, the platinum group isotopes were not supplied in large quantity by that method. Now the outlook is improved through the use of direct fluorination of the element by chlorine trifluoride fed into the arc chamber. Thus, for some elements the complexities and difficulties of separation have been solved, while for others problems still remain to haunt both operations and development groups as requests for more isotopes pour in.

The matter of calcium pumping deserves a short explanation. Typically, there are always problems in maintaining a sharply focused beam at high ion outputs, and these are worse with some elements than with others. Beams of calcium ions focus well as a rule, and this ease of maintaining focus is attributed to electron emission from the calcium-coated walls bounding the beam region. Following a hunch that such walls might benefit other separations also, we placed a calcium boiler below the beam adjacent to the ion source. When introduced into the beam in this manner at a rate of 2 to 3 grams per hour, the calcium produces benefits that are astonishing: the output of collectable ions is frequently doubled or even tripled, the beam stability is vastly improved, and the pressure of the system is reduced to minimal values by chemical reactions and by the gettering action of the deposited calcium. In fact, at high throughputs with halide charges, the calcium has a pumping capacity approaching that of several oil diffusion pumps 20 inches in diameter. Few such benefits are free; the use of calcium in this manner requires special handling to avoid fires and minor explosions. Thus we use caution, ventilate well when needed, and reap the benefits of a calcium-pumped system by providing better isotopes at a lowered cost.

In operating a calutron we are generally able to maintain the expected fixed focal position for the beam, yet we have also had beams take periodic excursions of up to ± 6 inches from the reference slot with a fixed magnetic field and accelerating potential—a condition we call "low-pressure tail wagging." The condition appeared to be caused by deficiencies in space-charge neutralization, and J. A. Martin discovered that it could be suppressed in large measure by "beam boundary plates." These are clean metal plates mounted close to the ion trajectories and parallel with them. The most effective design (the work of E. D. Shipley) included some 20 opposing plates comprising a pair of crescents that follow the ion paths, each set of subplates connected to its neighbor with an inductive coil and a capacitor. However, the necessity for routine washing and servicing dictated the use of a simplified design which consisted of two opposing crescent-shaped solid plates at ground potential. When calcium pumping was introduced the boundary was positioned ½ inch from the edge of the beam. This arrangement has virtually eliminated the occurrence of tail wagging, and has to some extent minimized the appearance of high-frequency random hash as the system is pushed closer to the point of maximum output.

These and other advances in technology were matched by increasing demands on our skills. It seems to me that I can measure my time at Oak Ridge in terms of isotopic purity increases. The isotopic distribution of the various elements in nature is such that about 50 percent of them are less than 15 percent abundant. In 1946, when the program was first started, a low-abundance isotope that was enriched to 90 percent was received with great enthusiasm by investigators. Shortly afterward, they wanted 95 percent. Later, high purity was thought of in terms of 98 and 99 percent, and finally our customers began manipulating the decimal point. The following stories will illustrate the present state of the art.

High-purity ^{233}U was needed for measurements of its fission-to-capture ratio—information fundamental to the concept of thermal breeder reactors. Uranium-233 does not occur naturally, and it has to be made in a reactor. The reactor, however, also makes ^{232}U. The investigators thought it was desirable to have the ^{232}U content in the sample reduced to between 1 and 10 parts per billion. From source material provided we collected 50 g of ^{233}U containing 4 ppb of ^{232}U. A check was made and the investigators called back and said they had erred in their calculations—the ^{232}U content was about eight times too high. A second separation reduced the ^{232}U content to between 0.1 and 0.2 ppb and the physicists were satisfied.

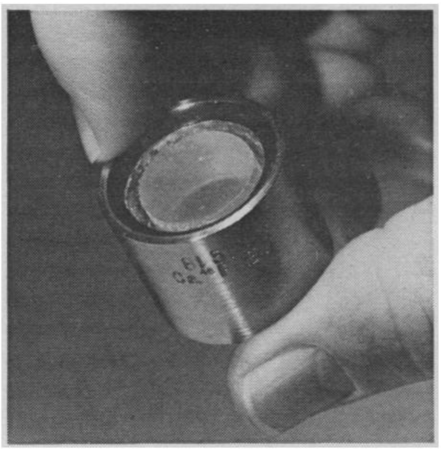


Fig. 6. Calcium-48 fluoride crystal used for an experiment in double beta decay.

Another example concerns a request for a 10-g sample of ^{48}Ca to be used in looking for double beta decay. Calcium-48 is 0.185 percent abundant in nature, and we had collected about 12 g of 96 percent ^{48}Ca and chemically purified it for the Harshaw Chemical Company to grow a calcium fluoride single crystal. A beautiful crystal (Fig. 6) was grown and sent to one of the national laboratories for the experiment. Two hours after the experiment started, the investigator called to say that the crystal was dirty and he would like to send it back and have it chemically purified. When asked what the contaminant was, he said "uranium," and added that it had at least 0.7 part per million. I mentioned that when we got anything to a chemical purity of 0.7 ppm we usually thought we had done pretty well, and asked him to hold the crystal until we could make a study. Describing the situation to G. Rogosa of the AEC Division of Research, I remarked that anything no larger than one's thumb and worth a quarter of a million dollars must be usable more than once; would he find another use while we determined whether we could improve the purity? He agreed and did find another customer for the crystal for a similar experiment, and we set about to see what we could do. In a second trial, we were able to get the uranium content in the material down to 0.5 ppb, had a new crystal grown, and sent it to the investigator; he looked at it, called back and said that it must not have been uranium—it must have been radium.

These stories are related not as a criticism of investigators who request such purity, but as an index of the level at which present-day investigators are working. One of the nicest aspects of this program is the tolerance of our customers and the appreciation they express for the materials that are made available. The separations group takes the job of providing enriched materials seriously and is acutely aware that experiments may fail because of calutron performance rather than the quality of the investigator's work. One of the shift supervisors expressed it best when he said, "For most of us, separating isotopes is not a job but a way of life."

One of our largest orders came in 1964, when the National Aeronautics and Space Administration requested the separation of 2500 g of tungsten-184 (natural abundance 30.6 percent) for use in developing nuclear propulsion for space. We equipped eight separators with double ion sources and receivers. (This was not an innovation; during the war some of the alpha separators had quadruple sources and four sets of receiver pockets working in parallel in a separator tank.) This is a tricky way to operate; the ion paths cross at the 90° deflection region and the two sets of beams are likely to interact, causing instability. Here our experience with the boundary plates was indispensable; they stabilized the beams and enabled us to complete the separation at a cost well below the estimate.

In addition to the 180° deflection separators that are mostly used, other focusing schemes known in mass spectrometry have been introduced. One is based on 255° deflection. Here one uses a magnetic field that is not uniform, but decreases with radius going outward from the center of the orbits. This gives focusing in the direction parallel with the magnetic field as well as normal to the field—the focusing is stigmatic. The main advantage comes from the greater dispersion that results from the longer path in the magnetic field, because this allows us to collect simultaneously all of the isotopes of a multinuclidic element such as tin, whereas with the 180° systems we could not provide adequate collector pockets for all isotopes. We now have six beta tanks equipped for 255° deflection.

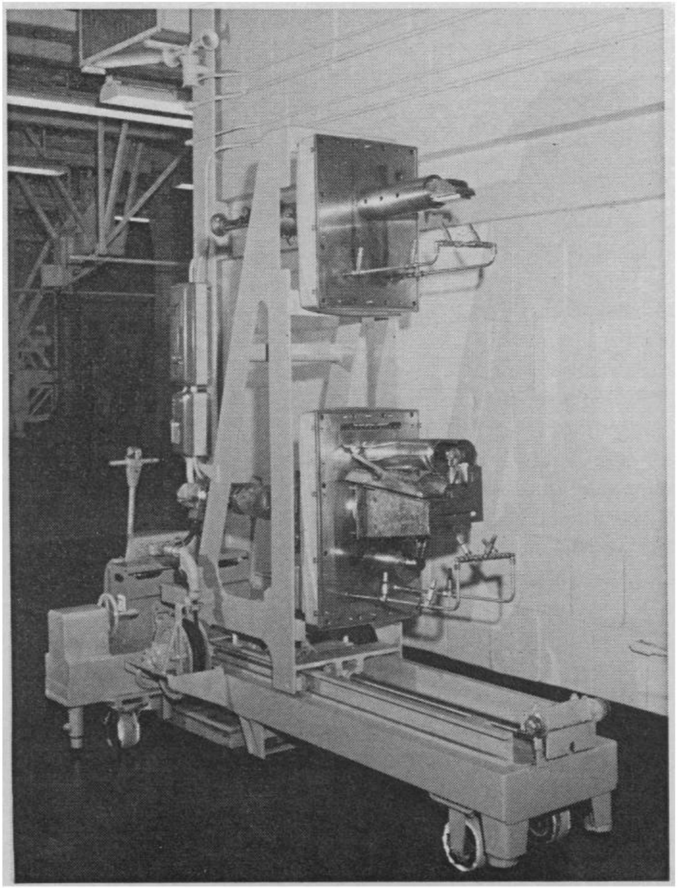
Another magnetic configuration is used in our 180° sector separator. Here the deflection is 180°, but the magnetic induction is so tailored that both source and receiver are 8 feet outside the magnetic boundary. This gives even higher dispersion (seven times that of the ordinary separator), and thus we use the machine when especially high isotopic purity is needed. For example, in a separation of dysprosium-156 for the production of ^{157}Dy for use in medical scanning of malignant organs, the assay was increased in a single pass from the natural abundance of 0.05 percent to a final abundance of 99.5 percent. Although the beam currents are low (3 to 10 milliamperes is typical), we are able to get usable quantities; also with this machine it is possible to place a decelerating potential on the receiver, so the ions enter the pockets with reduced energy, and this lessens many of the sputtering and heating problems. This feature makes the machine particularly useful in forming targets for accelerators.

Contained Facility for Radioisotopes

I have said little about the separation of the transuranium and other radioactive isotopes. Their availability in research quantities is just as important as that of the stable isotopes, and there is usually some point of special interest attached to each, inasmuch as their properties vary greatly from one to another. In reactor engineering their neutron capture cross sections have always been of interest, whether associated with fission product poisoning or the production of transuranics or for some other reason. In these days of breeding, more subtle properties are also of importance, such as the number of neutrons emitted per fission. In nuclear structure studies radioisotopes can be used to answer questions beyond the realm of the stable nuclides. As carrier-free tracers, they are in demand for radiochemistry, engineering, radiobiology, and medicine. As time goes on, the requirements on their quantity and purity become more and more exacting, again reflecting the increasing sophistication of experimental work. The quantities required range from a few milligrams to hundreds of grams.

In 1954 these needs were recognized at Oak Ridge, and a group comprising H. C. McCurdy, B. Harmatz, and F. N. Case devised a means for handling alpha-particle emitting materials in an electromagnetic separator. An enclosure is necessary because of the hazardous nature of most of the alpha emitters; our present facility houses eight calutrons that are used regularly for separations with thorium, uranium, and plutonium, and less frequently with americium and curium. Double containment is used; first-line containment is a glove box or the calutron itself, and second-line containment is the 8000-square-foot area, held at reduced atmospheric pressure, in which all processing is done. Two separators are assigned to such elements as thorium and certain plutonium and uranium isotopes, and a third separator has been modified to handle small amounts of ^{238}Pu, americium, and curium (Fig. 7). Isotopes which are less active (radioactive lead, bismuth, mildly active uranium isotopes, and so forth), but which should not be permitted to contaminate the stable isotopes area, are processed in the remaining tanks. Ten more separator locations adjacent to the present contained area are available for processing beta and gamma emitters such as curium and chemically toxic materials such as beryllium and osmium, should the program demand an expansion of contained facilities in the future.

When equipment is being washed or serviced, special clothing and masks are worn by personnel in the containment area. Care is taken to avoid bringing contaminants into the area as well as out of it because, for monitoring purposes, all alpha activity within the area is assumed to be attributable to the most dangerous element—plutonium. The equipment is transferred and serviced



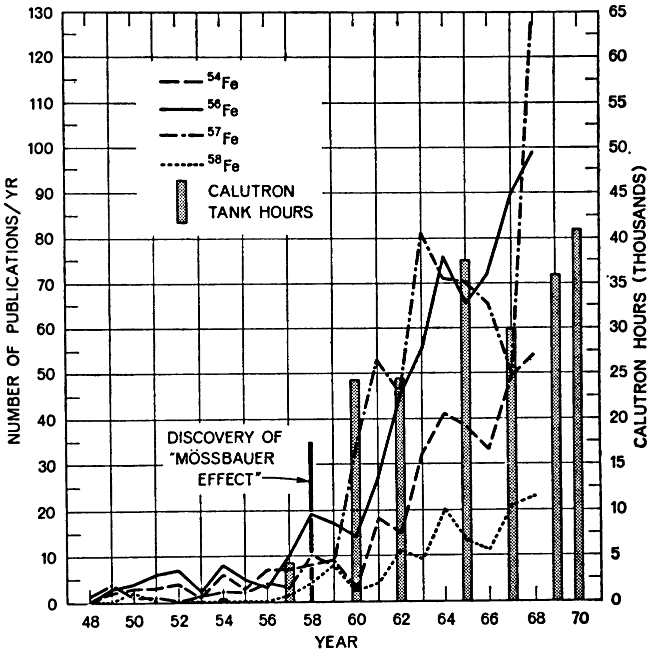


Fig. 7 (left). Apparatus used for the separation of isotopes of americium. Since the quantity of material used is small, both source (bottom) and collector (top) are smaller than standard. The dolly advances the mounting plates to the vacuum tank, where they are bolted on. The nozzles are used for cleaning with a spray of appropriate solvent while the equipment is inside its tank; thus, containment of the radioactivity need not be broken. Fig. 8 (right). Growth of research use of the iron isotopes as measured in scientific publications per year.

under plastic bags slightly below atmospheric pressure. The operations are time consuming, and procedures that take only minutes in stable isotope separations may require hours in the alpha-activity heavy element program.

The elements containing beta and gamma activity require special equipment to permit rapid handling and shielding protection, and in some cases in situ washing is necessary for equipment servicing (Fig. 7). A summary of our uranium and plutonium separations is given in Table 1. For the desired isotopic purity, some of the materials listed underwent two passes, and a few three passes. The feed materials from which most of these separations are made are produced in reactors, sometimes after years of irradiation.

Table 1. Uranium and plutonium isotope separations, 1960 to 1970. The amounts processed were: uranium, 27,400 g; plutonium, 7900 g.

Mass	Weight collected (g)	Assay range (%)
	Uranium	
233	220	99.99–99.9986
234	16	30.86–94.49
235	530	98.64–99.9988
236	280	85.07–99.996
	Plutonium	
238	5	99.48–99.9988
239	232	95.4 –99.999
240	238	97.03–99.993
241	120	83.31–99.997
242	360	81.45–99.987
244	3	0.61–99.06

Some Interesting Uses

Usually a particular separation is made to fill an order for a particular isotope, and the cost is assigned to that isotope. The other isotopes that are collected at the same time are prized byproducts, and they are held in reserve because of their potential value. Their market prices have to be somewhat arbitrary; they are determined in consultation with the Atomic Energy Commission. In some cases their potential values have been dramatically realized. I mentioned the 1948 separation of ^{202}Hg for W. F. Meggers; at that time we also collected ^{196}Hg, with a natural abundance of 0.15 percent. For over 15 years we had 300 milligrams of ^{196}Hg in inventory and no one had an active interest in it. Then someone realized that it could be placed in a reactor to make ^{197}Hg, and that ^{197}Hg has radioactive properties peculiarly well suited to the location of brain tumors. Since then ^{196}Hg has become one of the most sought-after isotopes. The history of ^{98}Mo is like that of ^{196}Hg—very little interest for about 20 years, and then a great demand (over 1.5 kg has been produced since 1965), again for medical scanning.

The case of calcium has different aspects. In early separations, the common isotope ^{40}Ca was placed in our sales inventory at 25 cents per milligram. Since then, including the big separation for ^{48}Ca, we have collected over 30 kg of about 99.99 percent ^{40}Ca, but it differs so little from natural calcium (96.96 percent ^{40}Ca) that it looked as if a use for it might not arise. However, an ingenious application in medical studies has recently appeared on the West Coast. If you feed a patient with foods containing ^{40}Ca, you can reduce the amount of ^{48}Ca in his blood; in fact, after a while the only ^{48}Ca in the blood must come from the reservoir in the bones. Thus, the rate of calcium loss from the bones can be measured by following the ^{48}Ca concentration in the blood, without affecting the overall chemical metabolism at all. We recently shipped 200 g of ^{40}Ca for use in this research, which, incidentally, is supported by the National Institutes of Health.

Iron provides further examples. Iron-56 is the abundant isotope (92 percent in nature). In early years we had separated iron primarily for the isotope of mass 58 (0.3 percent), which could be activated to ^{59}Fe and then used in a variety of blood studies in medicine, and in engineering research (on piston wear, for example). In doing so, we accumulated lots of ^{56}Fe and also an inventory of ^{57}Fe. Then two things happened. First, in 1958, the Mössbauer effect was discovered, and ^{57}Fe became of prime interest to physicists (Fig. 8). Second, in verbally advertising the availability of the ^{56}Fe, I discovered an investigator who said he could perform an experiment with 7 kg of ^{56}Fe, but it would be much better with 15 kg. This has to do with neutron shielding. The neutron cross section for iron has energy regions ("windows") where it drops nearly to zero; hence neutrons of those energies penetrate easily. This is important in nuclear reactor shielding, because iron is a common shield material. Iron-56 has such a window for neutrons of energy 24 kev. Thus two uses arise: in shielding studies, and to produce a beam of pure 24-kev neutrons simply by placing a plug of ^{56}Fe in the neutron beam from a nuclear reactor. A strong neutron beam of this energy is just about impossible to produce otherwise. We have processed 15 kg of our ^{56}Fe to pure iron for these applications. Iron-54 has somewhat similar uses, with its neutron windows at different energies. Thus, there is now a market for all of the iron isotopes.

In addition to outright sales, the separated isotopes can, under some circumstances, be obtained on loan. The Atomic Energy Commission maintains a Research Materials Collection, of which we are the custodians. Loans can be arranged from the collection when the material is not to be destroyed or isotopically contaminated. Details

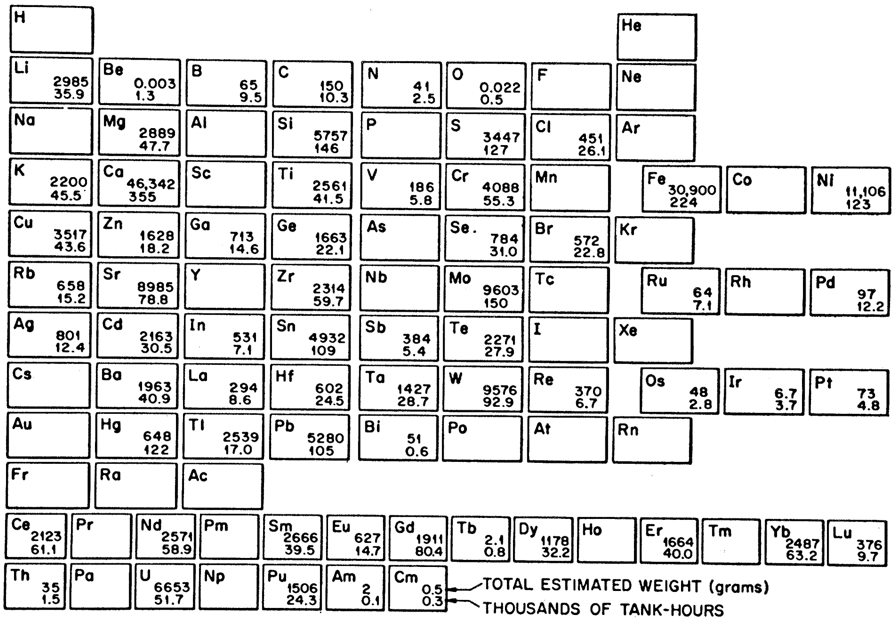


Fig. 9. Summary of isotope separations through 1972.

Table 2. Distribution of electromagnetically separated stable isotopes. Loans are made from the Research Materials Collection (*8*).

Fiscal year	Sales				New loans (equivalent value)
	Domestic		Foreign	Total	
	Non-AEC	AEC			
1966	$309,000	$172,000	$295,000	$ 776,000	$3,500,000
1967	425,000	265,000	297,000	987,000	6,600,000
1968	386,000	192,000	436,000	1,014,000	3,256,000
1969	373,000	203,000	523,000	1,099,000	5,780,000
1970	378,000	144,000	557,000	1,079,000	4,313,000
1971	262,000	102,000	559,000	923,000	7,400,000
1972	346,100	147,500	610,900	1,104,400	4,488,000

on both sales and loans are available in our catalog on enriched isotopes (*8*).

Figure 9 gives an overall summary of the separations since 1966, and Table 2 gives a summary of sales and equivalent dollar values of new loans. The steady growth of foreign sales shown in Table 2 may soon be affected by the entry of the Soviet Union into the European market.

An account of our output is incomplete without mention of the isotopic samples that are made by impingement, or implantation. Here the calutron beam is allowed to strike an appropriate substrate (usually a metal) and the ions (300 ev to 40 kev) bury themselves shallowly in the material and are left there. These isotopically separated samples are useful mainly as accelerator targets for experiments in nuclear physics and chemistry. The isotopes of the rare gases (stable or radioactive) are collected in this way. Since the targets are usually small (say, 1-cm squares), we can make them by the dozen at a cost of only a few dollars each. They are made according to the specification of the customer, and I estimate that during the course of our operations we have made more than 20,000 of these implantation samples for research uses.

The Future

On the technical side, we are testing automatic controls on the separators. This was a recommendation of an AEC advisory committee, and we are starting with a trial system on the 180° sector separator.

The indications are that the variety of demands for the separated isotopes will continue to increase. A survey of the research papers in the journal *Nuclear Physics A*, for example, shows that in 1966 there were 386 experimental papers, and 39 percent of them depended on the use of enriched isotopes, while in 1971 the numbers were 574 papers and 62 percent. The increase was uniform in the intervening years.

If one looks at the numbers of shipments made per year, one sees an almost exponential increase from 100 per year in 1946 to over 3500 per year in 1966. Since then the number has leveled off near 3600 per year (the highest was 3947 in 1969) and the trend has been toward somewhat larger shipments—hence the dollar-value increase since 1966 shown in Table 2.

Summary and Conclusions

In 1960 I attended a European conference on isotope separation, after which I visited the Niels Bohr Institute in Copenhagen. A staff member there ventured the opinion that the separation of isotopes will be first on the list of important contributions to the peaceful uses of the atom when the Atomic Energy Commission's memoirs are written in the year 2000.

In 1968 the AEC Division of Research contracted with the National Research Council of the National Academy of Sciences to conduct a review of the AEC program for the separation of stable isotopes by electromagnetic and thermal diffusion methods. This ad hoc panel comprised seven scientists from the fields of chemistry, classical physics, geochemistry, geophysics, medicine, and physics. In their final report on national uses and needs for separated stable isotopes (*9*), they referred to the store of separated isotopes as a "real national asset that attains increasing value as science and technology develop" and recommended "continuation of the program as a national resource of great value to the United States."

Later, in a discussion of this report with A. M. Weinberg, J. Koch, himself a pioneer in electromagnetic isotope separation and member of the Danish Atomic Energy Program, said he would correct the statement that the Oak Ridge electromagnetic facility is a "national asset" to read "international asset."

From my narrow viewpoint after an extended and complete engrossment with this program for so many years, it is gratifying to learn that such men as those mentioned above share my belief that the work has indeed been worthwhile.

References and Notes

1. The apparatus was called a calutron, a contraction of California University Cyclotron, because it made use of the magnet from one of the University of California cyclotrons. Alpha and beta identified the calutron types on the basis of first- and second-pass separations. The alpha calutrons (48-inch radius) enriched the ^{235}U from its normal abundance of 0.7 percent to about 15 percent, and the beta (24-inch radius) from this purity to about 90 percent. The radius figures refer to the size of the semicircular orbits traversed by the ions.
2. A. J. Dempster, *Phys. Rev.* **11**, 316 (1918).
3. J. Classen, *Physik. Z.* **9**, 762 (1908); *Ber. Physik. Ges.* **6**, 701 (1908).
4. A. O. Nier, E. T. Booth, J. R. Dunning, A. V. Grosse, *Phys. Rev.* **57**, 546 (1940); *ibid.*, p. 748.
5. For two isotopes, the enrichment factor is defined as

$$(A_1/A_2)_{\mathrm{final}}/(A_1/A_2)_{\mathrm{original}}$$

where A is abundance, usually expressed in atom percent.
6. G. Sidenius and O. Skilbreid, in *Electromagnetic Separation of Radioactive Isotopes*, M. J. Higatsberger and F. P. Viehböck, Eds. (Springer-Verlag, Austria, 1961), pp. 243–249.
7. J. A. Swartout, G. E. Boyd, A. E. Cameron, C. P. Keim, C. E. Larson, *Phys. Rev.* **70**, 232 (1946).
8. The catalog, *Research Materials, Separated Isotopes and Radioisotopes, and Special Preparations*, is available free from Oak Ridge National Laboratory Isotope Sales, P.O. Box X, Oak Ridge, Tennessee 37830.
9. "Ad hoc panel report on the national uses and needs for separated stable isotopes," an edited version of the National Research Council report, *Isotop. Radiat. Technol.* **7** (No. 4), 363 (1970). This publication is available from the Government Printing Office, Washington, D.C.