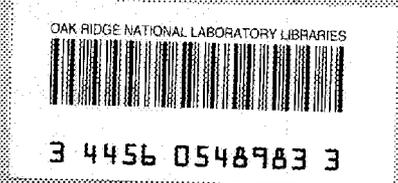


OAK RIDGE NATIONAL LABORATORY
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ISOTOPES DEVELOPMENT CENTER

RESEARCH MATERIALS COORDINATION AND PLANNING MEETING

NOVEMBER 14-15, 1963

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Sponsored by

ORNL ISOTOPES DEVELOPMENT CENTER

and

U. S. ATOMIC ENERGY COMMISSION

Compiled by

P. S. Baker

APRIL 1965

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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RESEARCH MATERIALS COORDINATION AND PLANNING MEETINGNOVEMBER 14-15, 1963

INTRODUCTION

This report includes most of the talks and discussion presented at the Research Materials Coordination and Planning Meeting held at Oak Ridge National Laboratory on November 14-15, 1963. Unfortunately, the last copy was not received until mid-September, so that the material may have lost some of its timeliness. Nevertheless, it is still probably worth recording - if to serve only as "minutes" of the meeting.

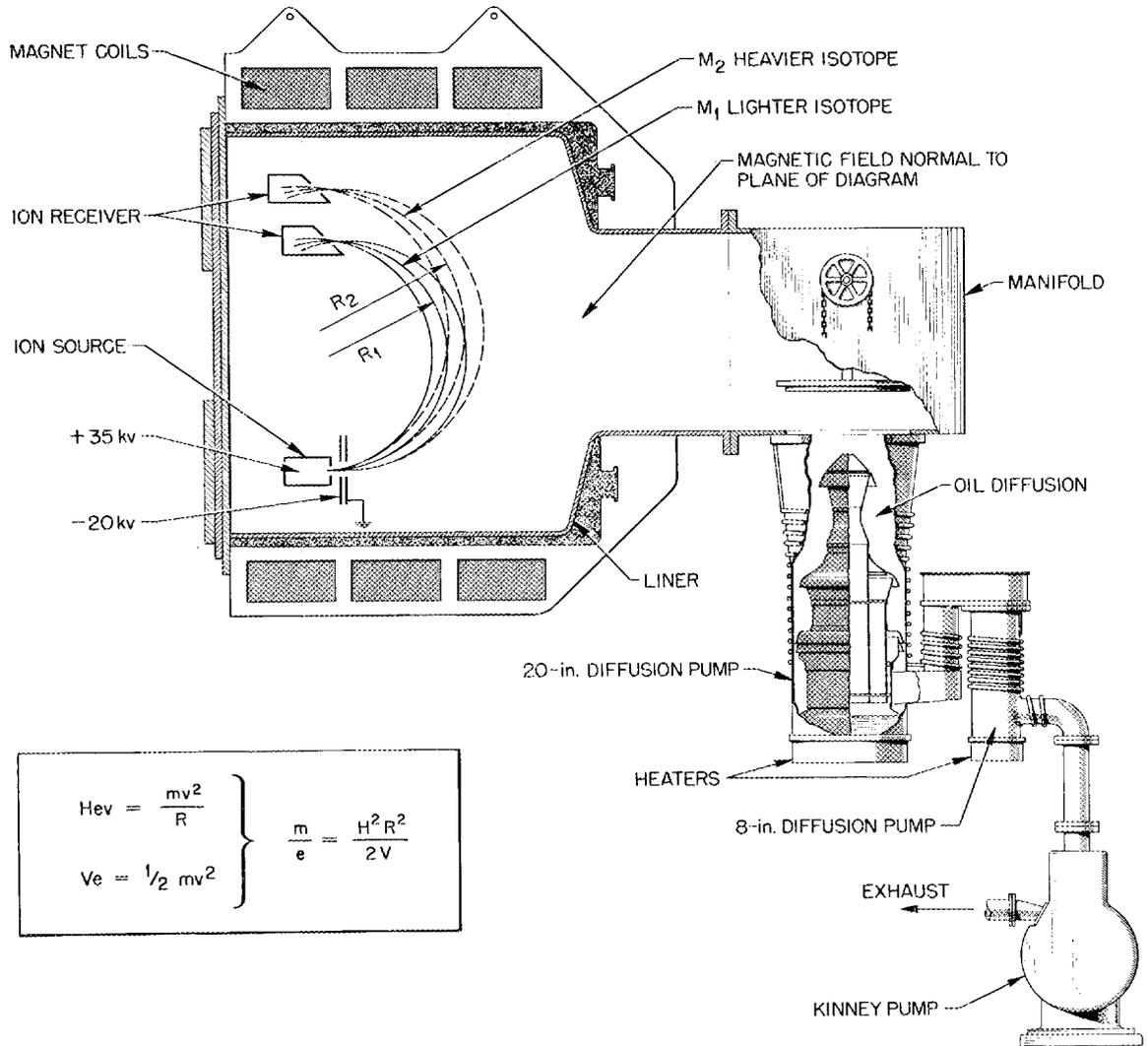
ELECTROMAGNETIC ISOTOPE SEPARATIONS

L. O. Love, ORNL

We who have responsibility for operating the electromagnetic isotope separation facility consider our position a favored one in that we are privileged to provide materials for people conducting experiments which will influence the design of nuclear instruments over the next 5 to 20 years. Users of separated isotopes have at their disposal--through proper channels--a multimillion dollar plant containing a total of 68 electromagnetic separators. What I am going to do in the next few minutes is to describe what these facilities are, what is being done with them, what it costs to operate them, and what might be done with them should the need arise.

The original electromagnetic separations plant was built to process one element to collect one isotope and as such was designed to operate at only one set of parameters. Today the facility stands as the most versatile isotope separating plant in the world. All isotopes of the chemically active elements appearing in the periodic chart have been collected and made available in usable amounts, and today six elements are routinely processed simultaneously. Forty of the separators assigned to the program are in continuous use.

Many people who are familiar with electromagnetically separated isotopes may not be familiar with the process itself. Figure 1 shows a schematic of the system and the basic calutron equation. For technical reasons it is more desirable to be able to vary the magnetic field from element to



Schematic of the Calutron

Fig. 1

element than to change any other operational parameter. Figure 2 shows one portion of the facility as it appears today. Originally there were 36 units contained in one common magnetic field, usually referred to as a "track". In order to provide flexibility, modifications were made which permitted this track to be segmented into four magnetically isolated systems, thus allowing four different magnetic field strengths to be used simultaneously. This allows concurrent separation of four different elements in a system where previously only one element could be processed.

A second track seen in the background of this same picture has been modified to magnetically isolate eight separators. This segment with necessary chemistry laboratories for processing alpha active materials has been enclosed to provide a double containment area for separating elements such as Pu, U, Th, and other elements with similar activity. The remaining separators in this second track are in various stages of renovation and could be activated for processing kilogram quantities of stable isotopes should the need arise.

What we have been able to accomplish can be illustrated by the following figures. Figure 3 shows our annual rate of collection for the past few years. It is interesting to note that from 1946 to 1959 we collected a total of ~20 kg of separated isotopes. In the next three-year period we collected twice this amount. We have recently completed our first year of processing the alpha active element plutonium in our new containment facility; Table 1 shows the results of this effort.

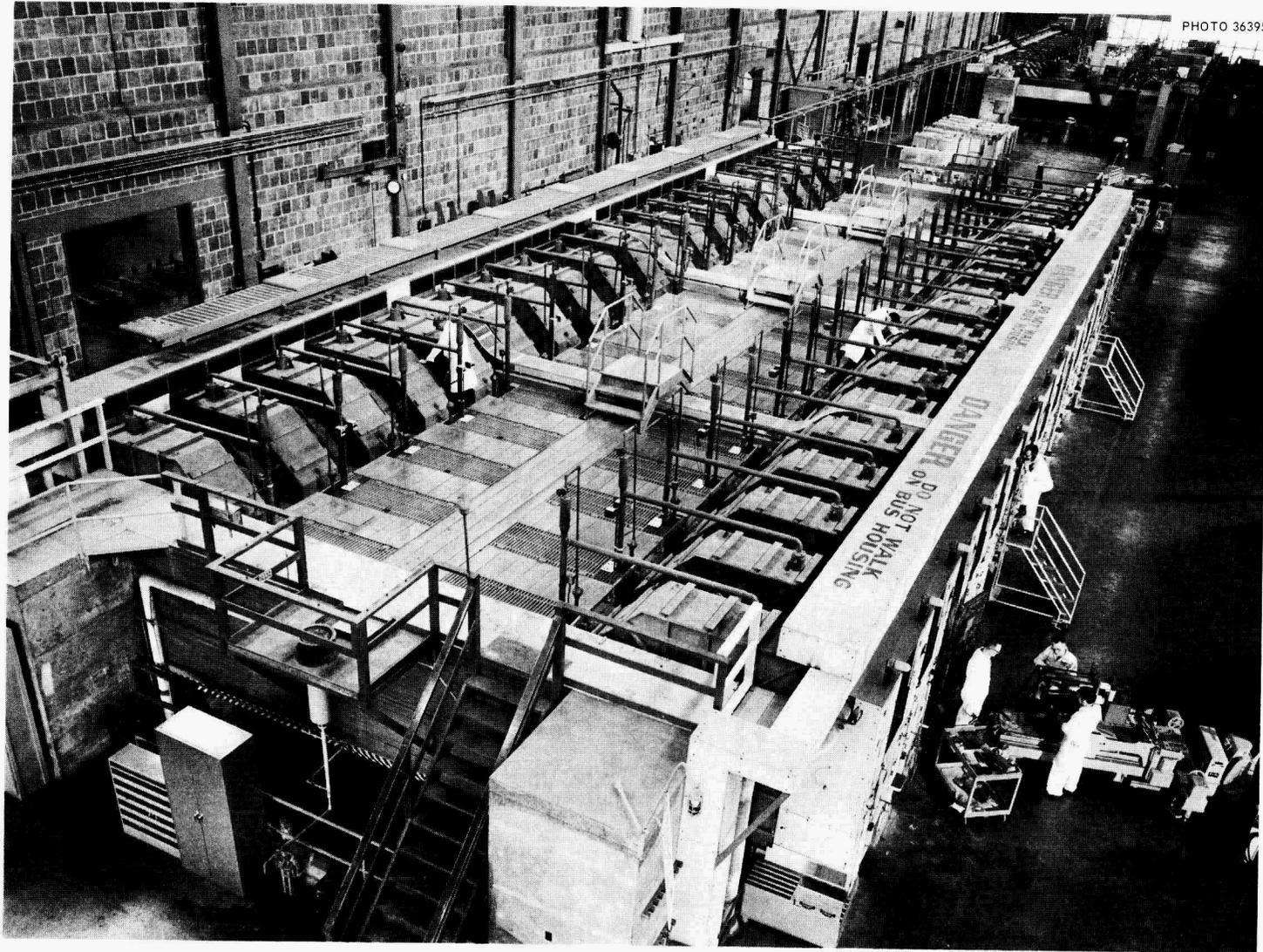


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Fig. 2. Portion of the Electromagnetic Isotope Separation Facility

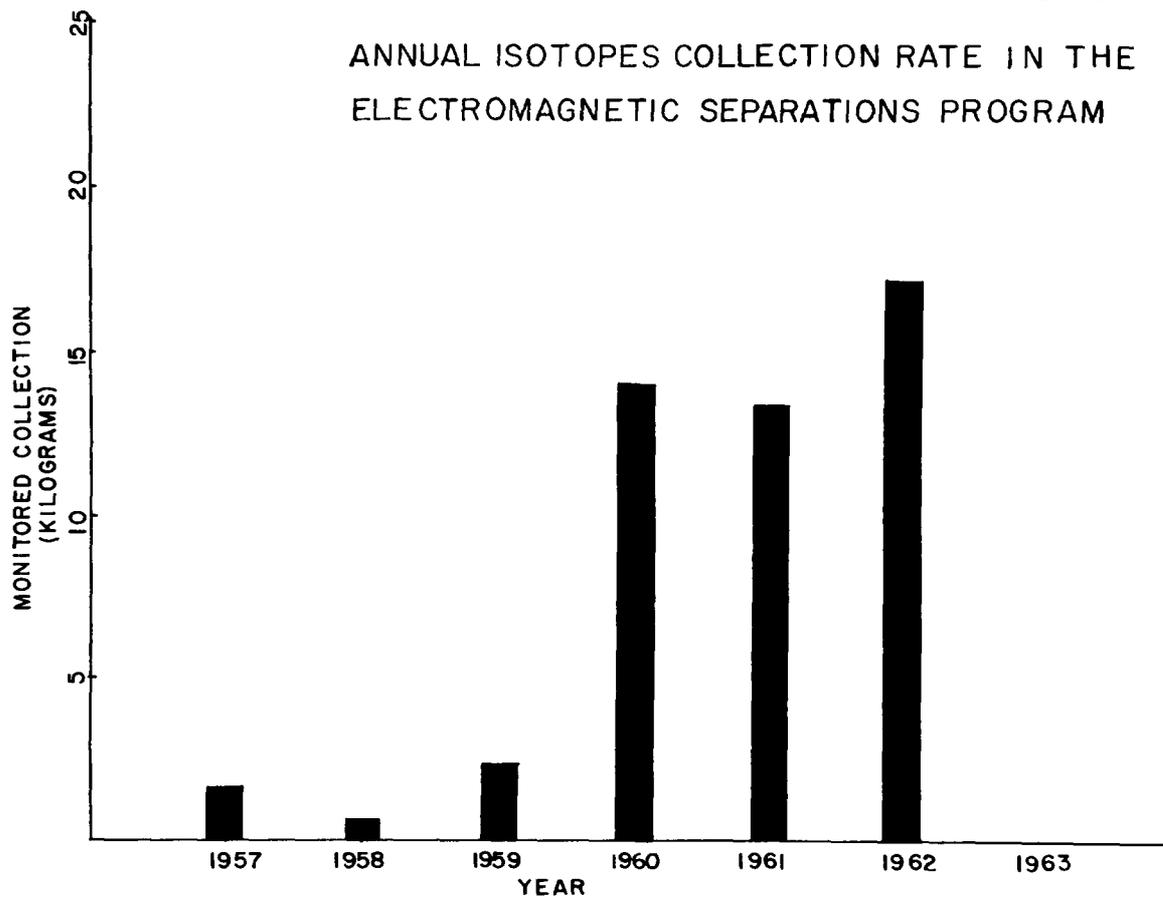


Fig. 3

Table 1Quantity and purity of collected isotopes
plutonium series "a"

Weight of isotope equivalent to monitored ion currents, g		Purity range, %
^{242}Pu	16.0052	84.22-93.76
^{241}Pu	32.2836	89.20-95.88
^{240}Pu	126.1935	97.63-98.96
^{239}Pu	71.7178	94.88-94.96

Figure 4 shows some enrichments which have been provided. In some instances this involves concentrating isotopes which occur in minute quantities in the feed material, at other times it involves reducing contaminants to minute quantities where the isotope is already in abundant supply in the charge material.

In addition to handling ordinary chemicals we sometimes process highly toxic materials. Figure 5 shows our approach in handling elements such as beryllium and osmium.

In 1947 the electromagnetic plant was shut down because of its inability to compete economically with the K-25 process. Today it is still an expensive business but there are two points worth noting.

1. The cost of a separator tank hour in 1947 when we were processing only one element for one isotope was ~\$10 exclusive of chemistry costs. Today when we are processing more than 50 elements and collecting more than 250 isotopes, the cost is still ~\$10 an hour and this now includes chemistry.
2. The electromagnetic process was not only the first process to collect ^{235}U , it is the first and only process to collect usable quantities of all the chemically active elements.

Figure 6 shows the effect of increasing the number of separators on the total cost of the program. In following the incremental cost line, it is seen that the important thing being changed in reducing the number of tanks is quantity of material collected. The total dollar cost is not

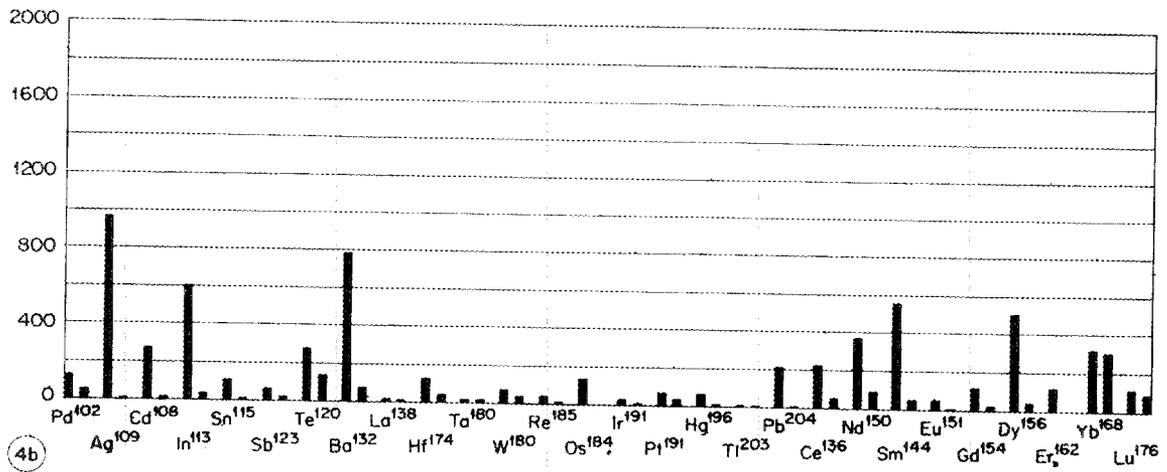
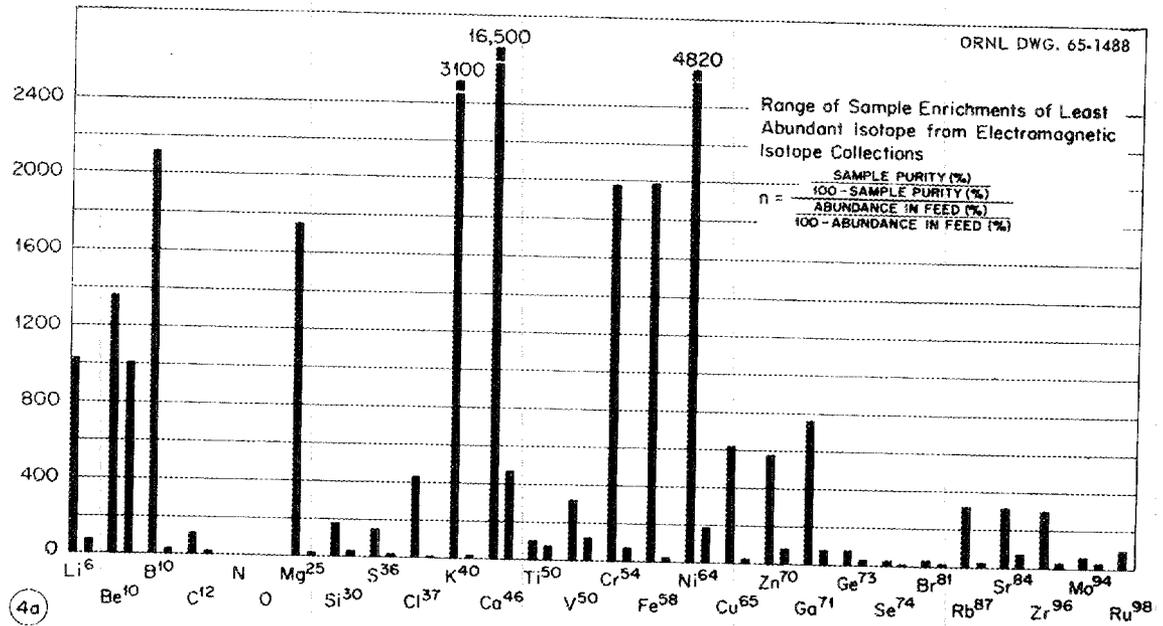


Fig. 4

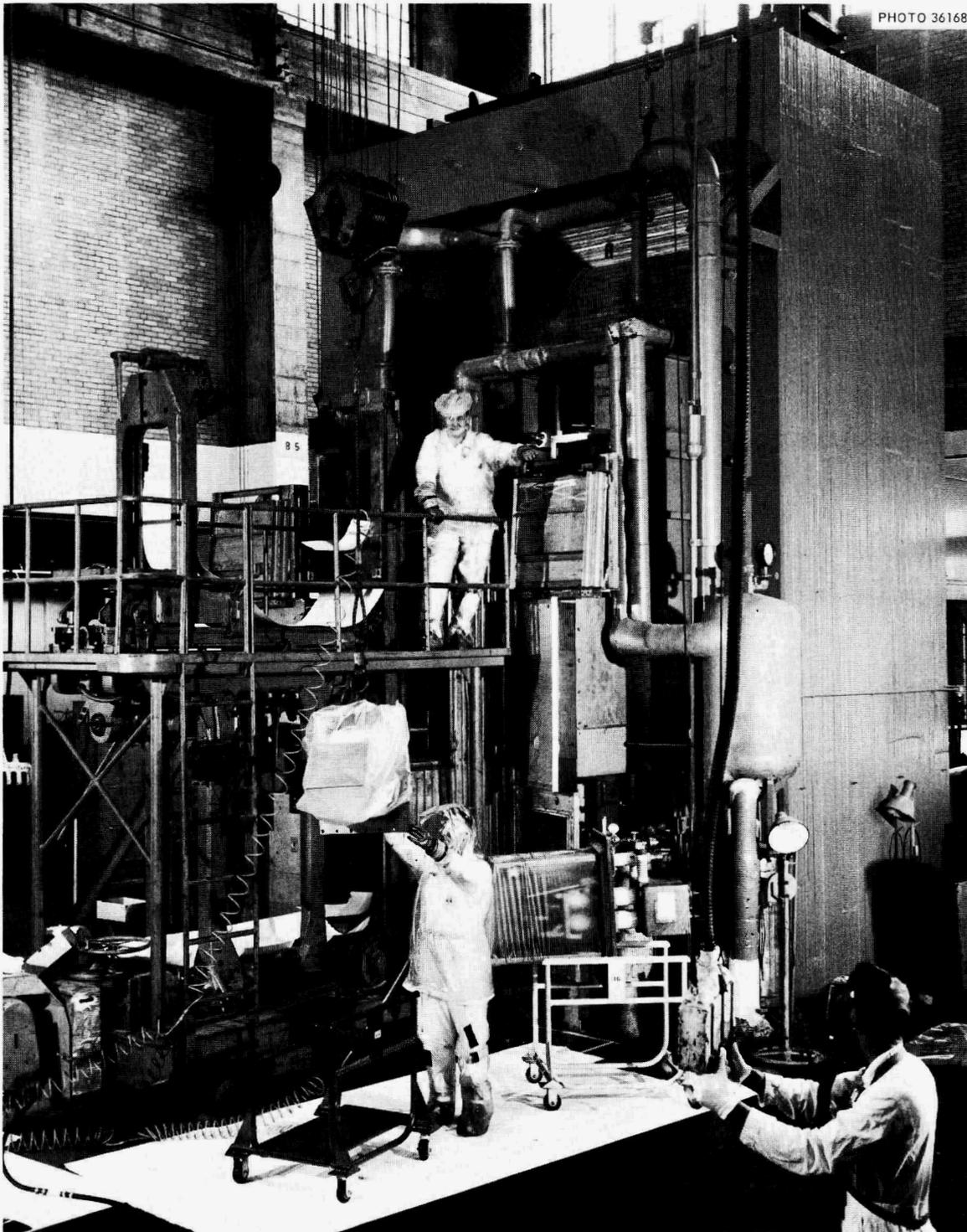


Fig. 5. Approach in Handling Elements such as Beryllium and Osmium

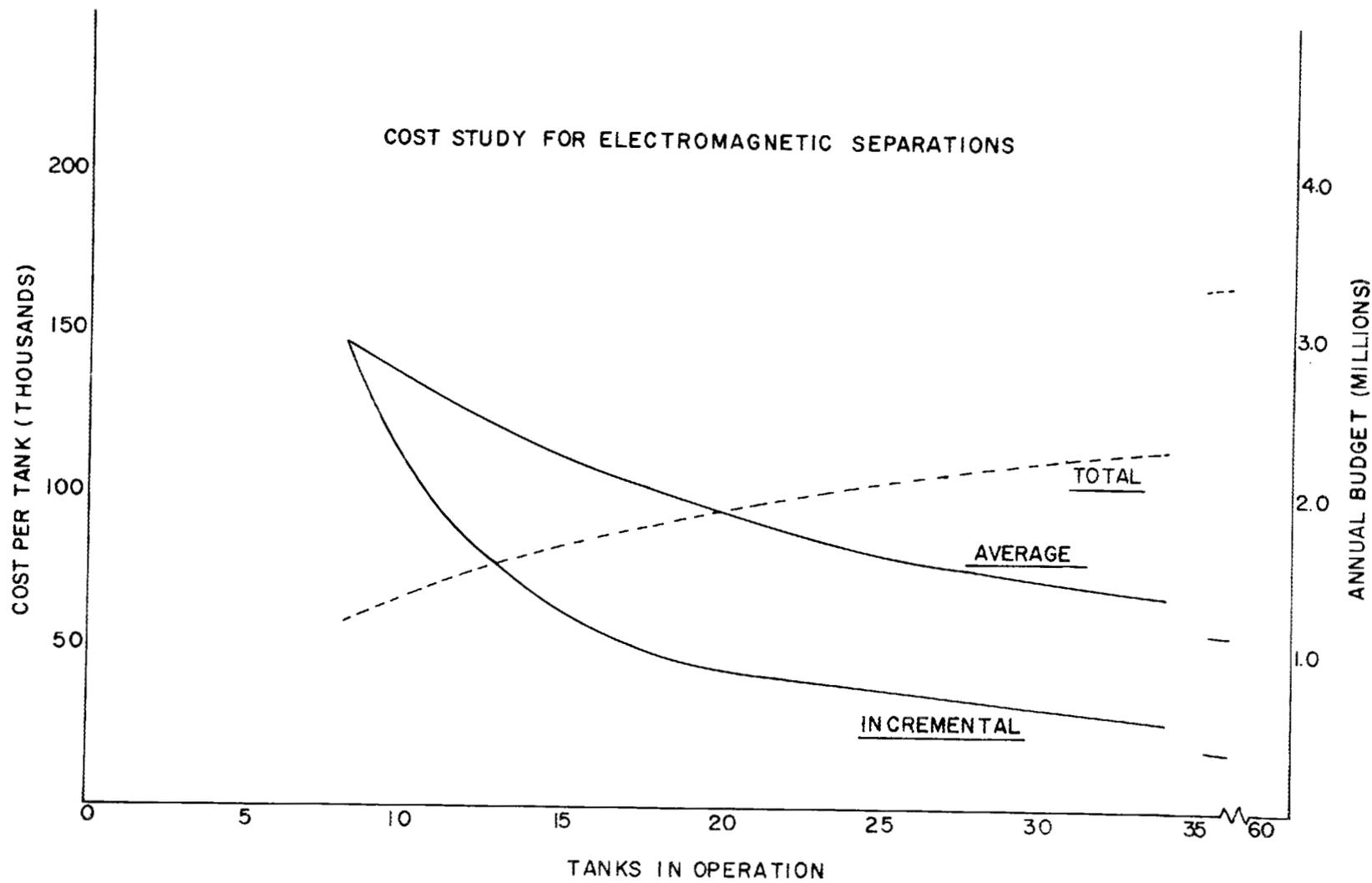


Fig. 6

affected appreciably until the zero point is reached. One effect of a larger scale operation is clearly shown in Fig. 7 which compares the cost and purity trends of a single isotope (^{48}Ca) since 1948.

Always of prime importance to any program are the people assigned to it. Figure 8 shows the distribution of personnel in the Department according to work or training. The group labeled "other" can be classified more in the general science category. Their work experience is specifically in the art of separating isotopes by the electromagnetic process, and this designation includes some people who have been with the program for over 20 years. Another item worthy of note is that 25% of the technical people in the Department are going to graduate school. A number of men are currently doing project work in connection with their theses, and several papers are in various stages of completion. We believe there are fundamental processes operating in the calutron which, when they are understood, will permit increases in purities and outputs far beyond those now experienced. We believe that development of these principles will come primarily from the group of people now associated with the electromagnetic separations program.

There is probably no other place in the AEC where a full-scale research program in such fields as plasma physics or ion propulsion could be established for such small capital outlay as could be done here; and should it ever become necessary to process one element to obtain kilogram quantities of a specific isotope, the entire 68 separators could be diverted on very short notice to achieve this objective.

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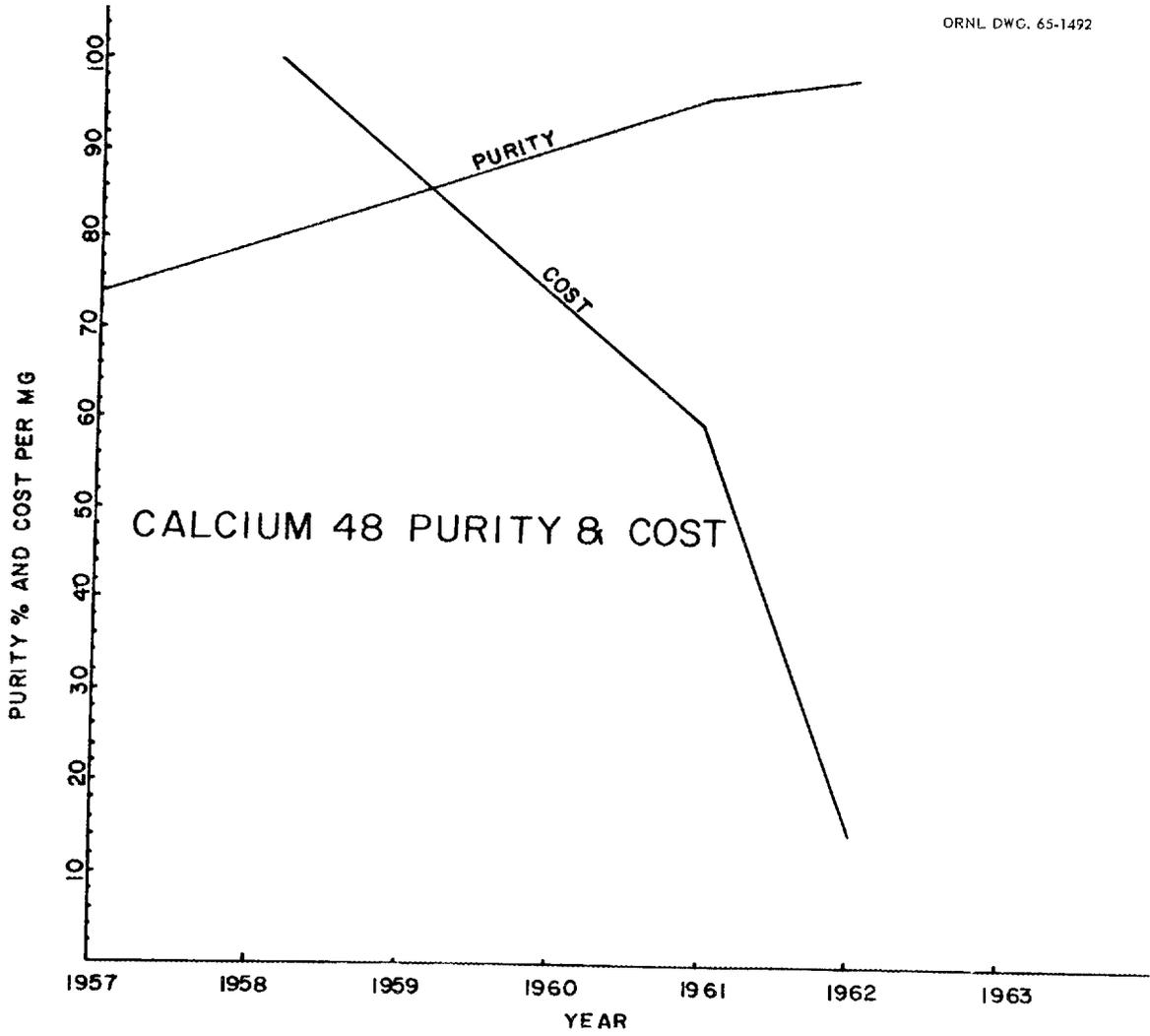
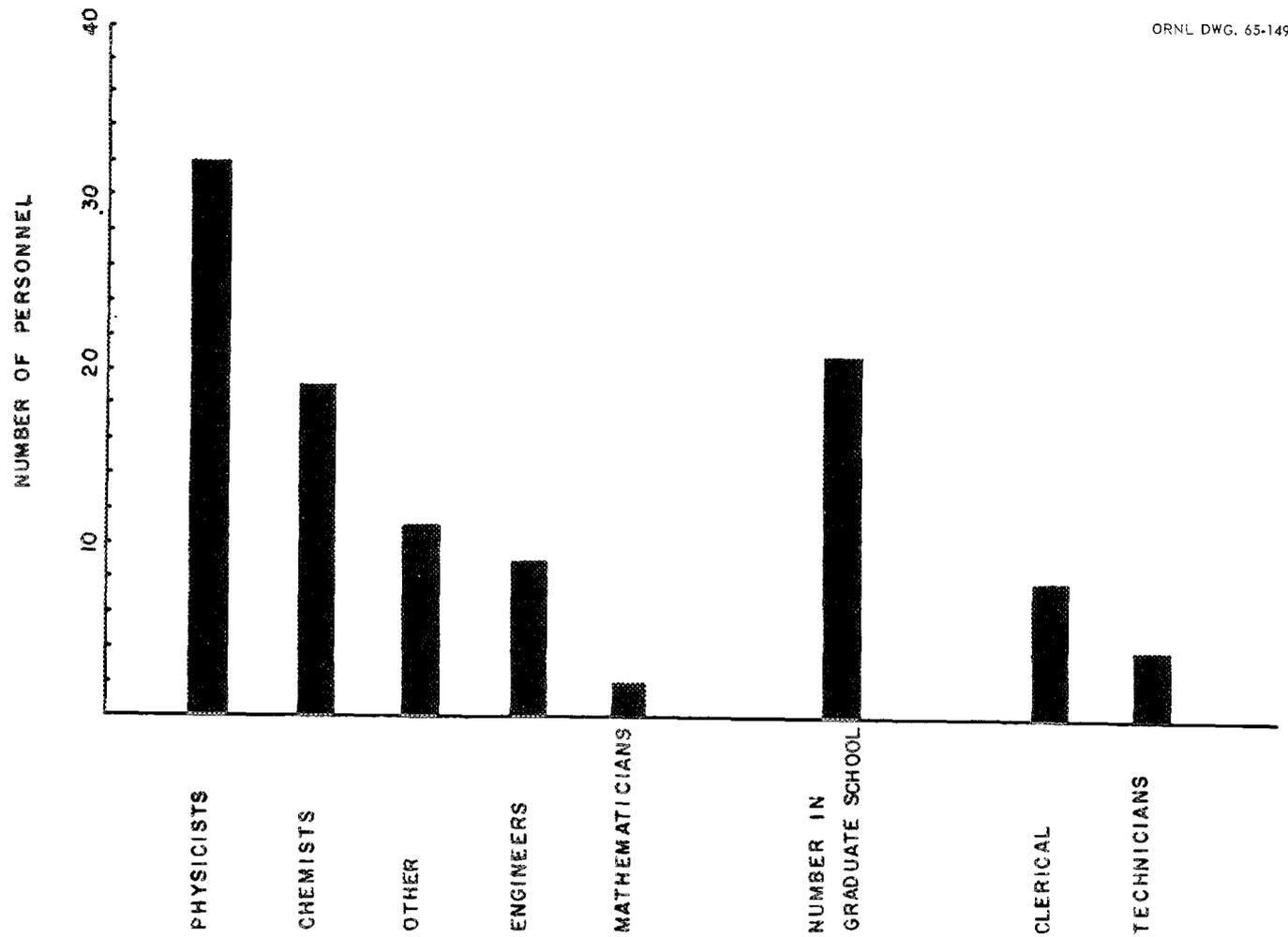


Fig. 7



PERSONNEL IN ELECTROMAGNET ISOTOPES DEPARTMENT

Fig. 8

THERMAL DIFFUSION

W. R. Rathkamp, ORNL

The Isotope Division of Oak Ridge National Laboratory is responsible for the production of stable and radioactive isotopes, not available from private industry, that are required by both commercial customers and government agencies. As part of this isotope production effort, the thermal diffusion program has the responsibility of making available enriched isotopes that either cannot be produced by other separation methods or are more costly when prepared by alternate means.

Most of our work in the past has been concerned with enriching various isotopes of the inert gases. Neon-20 has been increased from its natural abundance of 90.8% to over 99.99% concentration, and neon-22 has been raised from 8.9% to 99.9% in liter quantities on a routine basis. A smaller amount of neon-22 was prepared with an isotopic purity of greater than 99.999%. Both neon-20 and neon-22 have been sold in liter quantities to be used in the construction of improved lasers.

Argon-36 has been enriched from its natural abundance of 0.34% to 99.9% also in liter quantities.

The noble gases of higher atomic weight are somewhat more difficult to process since the unit separation factor that can be achieved decreases strongly as the mass increases. However, krypton-86 has been taken from 17.3% to greater than 99.9% for use as a wavelength standard, and krypton-78 has been enriched from 0.35% to 10% concentration to provide material for cyclotron bombardment experiments. As a by-product of the krypton-86

enrichment program, the feed material, depleted in mass 86 isotope, was used in the cyclotron production of rubidium-85.

The concentration of xenon-124 has been increased from 0.094% to over 4% in our newest operating project, and eventually a product containing 10% xenon-124 is expected. This material will be neutron irradiated in experiments to produce iodine-125.

Neon is composed of a mixture of three isotopes, of mass 20, 21, and 22, and argon similarly contains three isotopes, of mass 36, 38, and 40. The heaviest and lightest isotopes in any such mixture, such as neon-20 and 22, or argon-36, can easily be enriched to any desired concentration if a thermal diffusion system is constructed with sufficient length. The problem of increasing the concentration of a middle isotope, such as neon-21 or argon-38, is much more formidable. A cascade to enrich an isotope of other than the greatest or least mass in a mixture of three or more components may become quite complicated in arrangement, and require careful design in regard to length, width, and flow rates. Two such middle isotopes enriched in our laboratory are neon-21, presently available at 32% isotopic concentration raised from 0.26% abundance, and argon-38, enriched from its natural concentration of 0.063% to 23%. One use for argon-38 is in isotope dilution to facilitate mass spectrometer measurements in age-of-the-earth determinations.

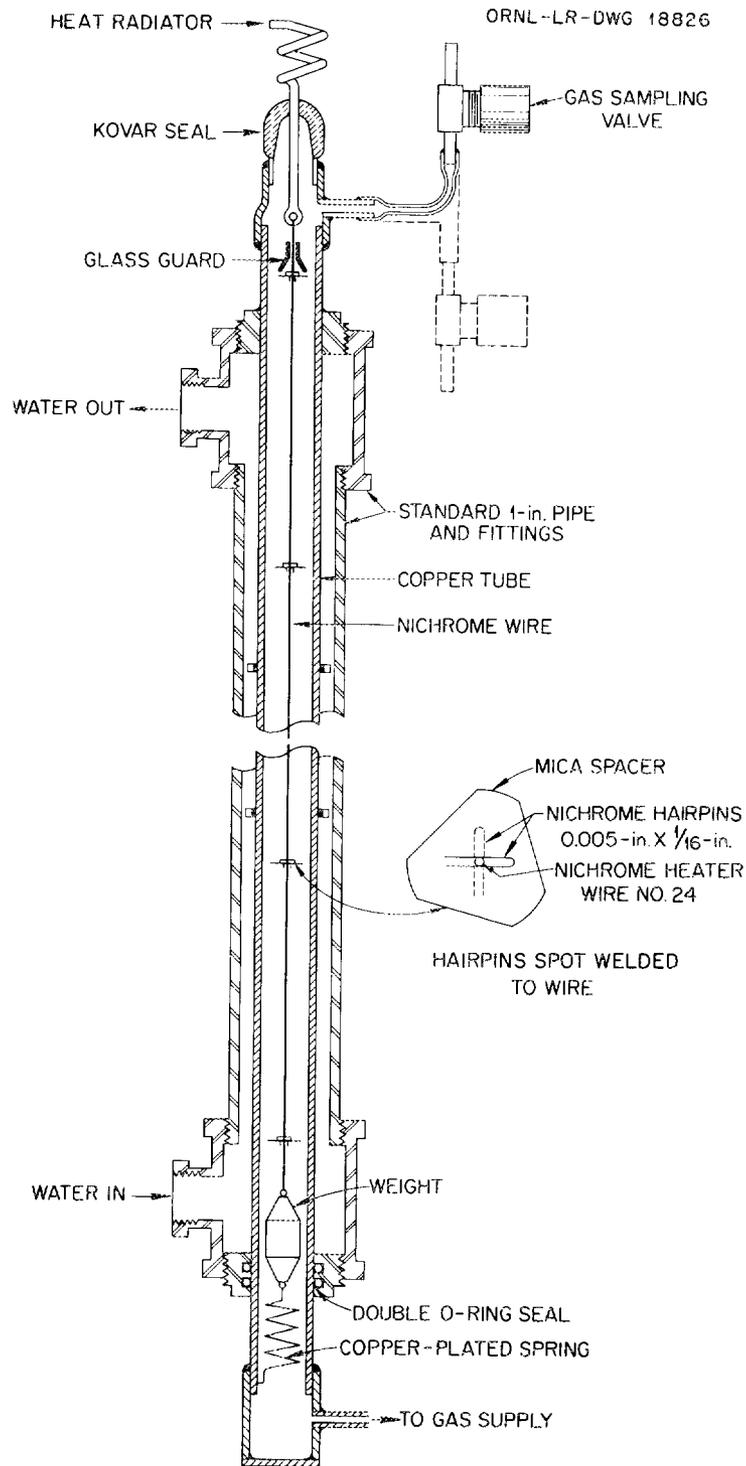
We are presently engaged in the engineering and construction of a cascade to enrich the radioactive fission product krypton-85 from 6% to 50% isotopic concentration. Material of this composition

is desired to build self-exciting fluorescent lamps for use in railroad signals and buoy lights, and in the construction of power sources for missiles and satellites. Such a thermal diffusion system must be designed for the ultimate in reliability, since maintenance work on a plant containing radioactive material will be difficult and must be held to a minimum.

Carbon-13 has been produced at a concentration of 20%, enriched from its natural abundance of 1.1%, using methane as feed gas. Thermal decomposition of the methane has prevented the achievement of higher isotopic concentrations, but recent experiments have shown that decomposition is very much less when aluminum sheathed heaters are employed in place of the stainless steel heaters previously used. The cascade will be rebuilt using aluminum heaters, and is expected to produce 90% carbon-13.

The thermal diffusion work in our laboratory started with the use of hot wire type columns as shown in Fig. 1. The heater consisted of a Nichrome wire 0.020" in diameter along the axis of a copper tube 7/16" inside diameter. The overall length of these columns was 21 feet. While much useful work was accomplished with this style of construction, trouble was experienced with frequent failure of the hot wire.

The columns were redesigned to use standard commercial tubular heaters to eliminate this difficulty. This type of column is shown in Fig. 2. These heaters not only proved to be more reliable in operation than the hot wire heaters, but could be operated from a 460 volt AC source of power, whereas the hot wires required a



Details of Hot Wire Thermal Diffusion Column.

Fig. 1

THERMAL DIFFUSION COLUMN CROSS SECTION

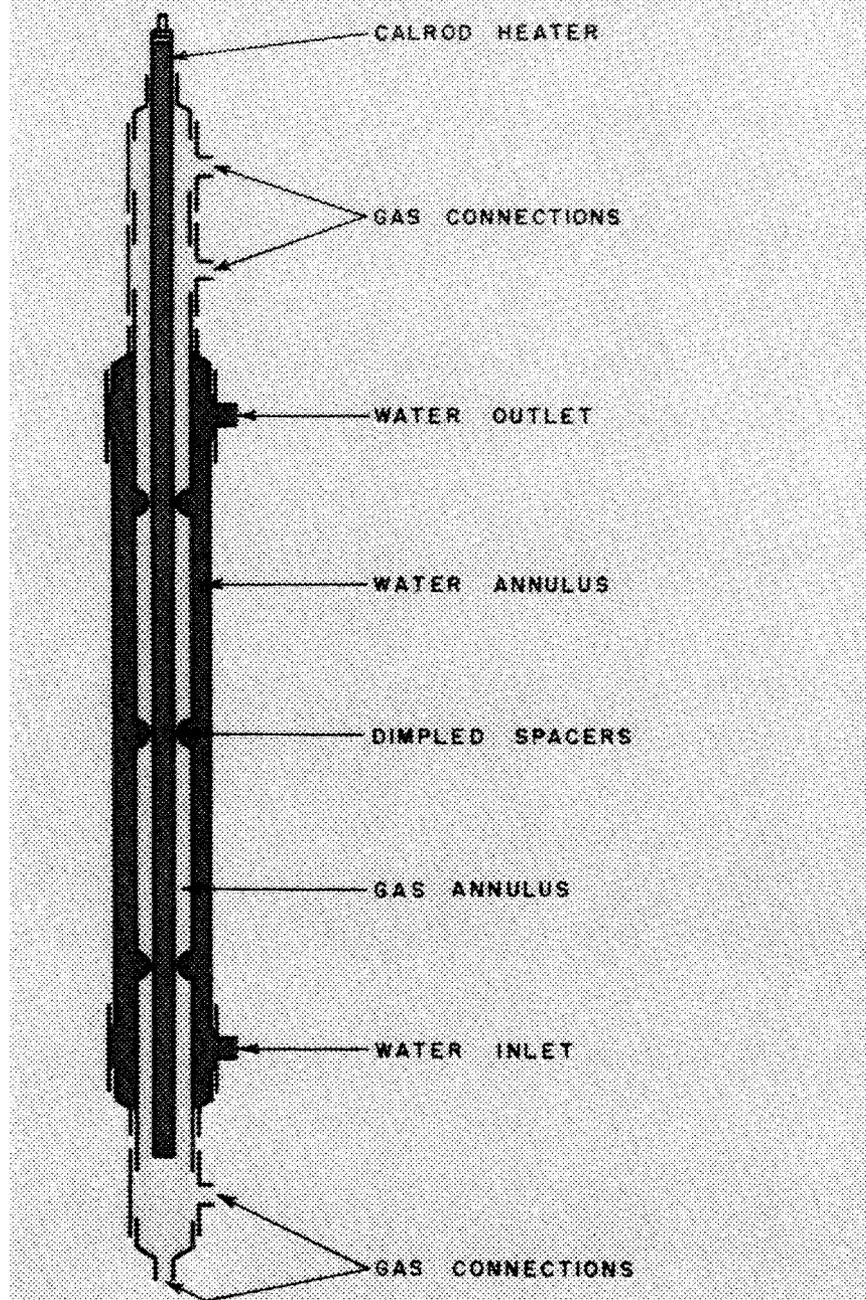


Fig. 2

direct current supply. The heaters are 0.440" in diameter and have an effective heating length of 7 feet. The column length can be doubled to give 14 feet of length if a second heater is inserted through the bottom end. Since column performance can be improved by using a smaller annulus and a higher gas pressure, the present columns have an annular gas space 1/8" wide. The tubular heaters are held along the axis of the tube by small dimples impressed into the cold tube walls.

There are 45 columns of this type in operation in our laboratory. Since this number of columns require a fairly large floor area, it appeared desirable to design columns of this same geometry in bundles enclosed in a common water jacket in order to conserve laboratory space. This construction is shown in Fig. 3. Thirty six columns each 7 feet long are enclosed in a shell 8" in diameter. We now have four of these bundles, totalling 144 columns, occupying approximately 14 square feet of space. Since all the columns are sealed individually with the flat head alternate arrangement, the individual columns can be interconnected externally to give any desired shape of cascade. Also, some of the tubes may be used for enriching one isotope at the same time other columns are being used to enrich a different isotope or element.

While the field for gaseous thermal diffusion is by no means exhausted, there are many elements containing isotopes of interest which do not have any suitable gaseous compounds. Some examples are stable calcium-46 and the radioisotopes cesium-137 and strontium-

THERMAL DIFFUSION COLUMN BUNDLES

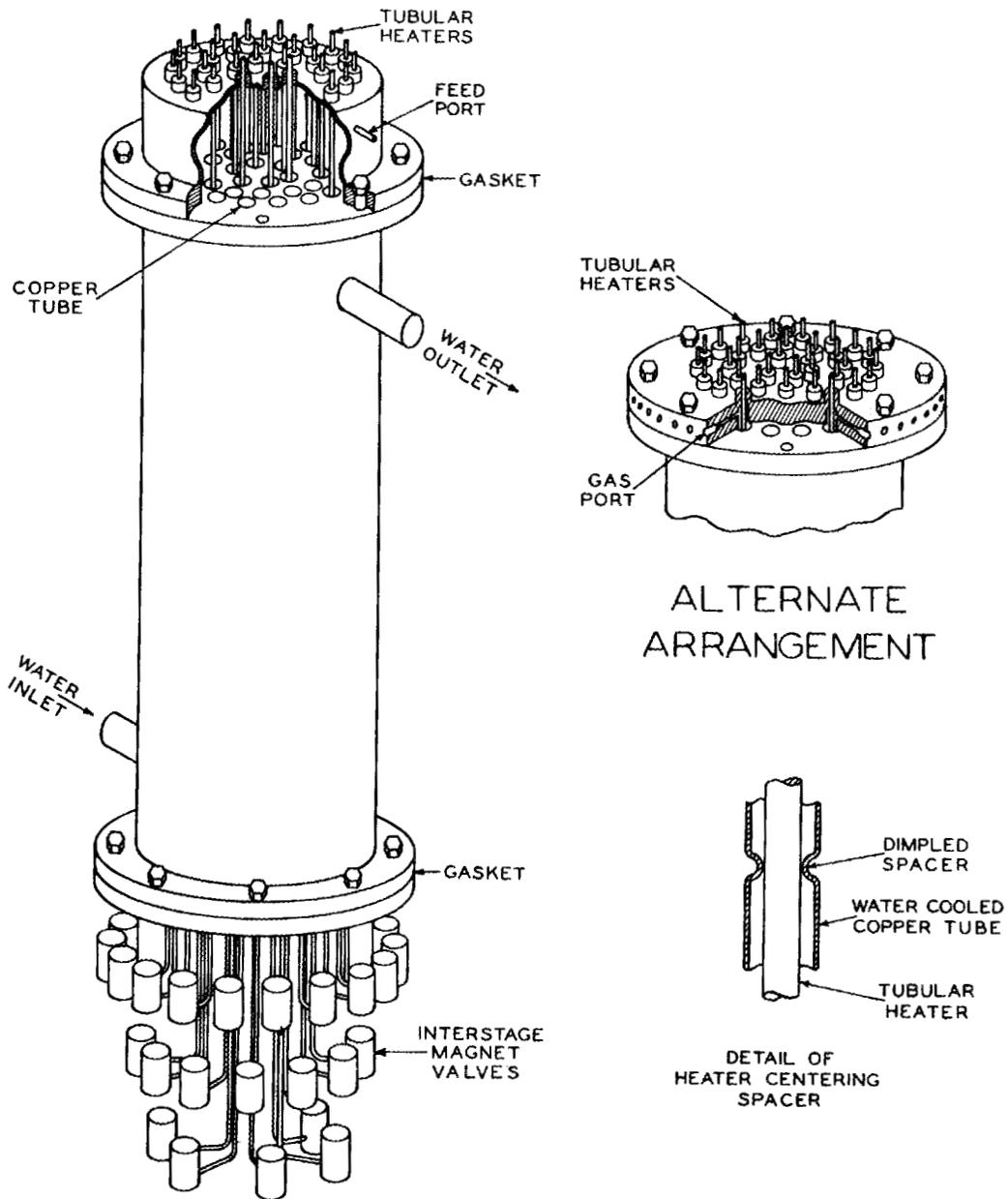


Fig. 3

90. If thermal diffusion in the liquid phase could be adapted to production systems, it would be possible to enrich many more isotopes than have been prepared to date. We would like to investigate thermal diffusion in either molten media or in solutions of elements to make available additional isotopes at greatest purity and minimum cost.

While the thermal diffusion process is in general not competitive economically with other methods of enriching isotopes, it is valuable for processing elements such as the noble gases for which few other isotope separation methods are available. Since the initial cost of installing a thermal diffusion system is quite low, the process is often economical for small scale operations involving any suitable element or compound.

Experience has shown that very little maintenance or operational attention is needed by a well designed thermal diffusion cascade. This fact may make the process desirable even on a larger scale for handling radioisotopes where equipment maintenance and remote operation are important factors.

ISOTOPES OF HYDROGEN, LITHIUM, BORON,
NITROGEN, OXYGEN

J. S. Drury, ORNL

Since 1947 members of the Chemistry Division, ORNL, have been interested in separating isotopes of the light elements. During this time we have developed methods for separating, or have actually separated, isotopes of hydrogen, lithium, boron, and nitrogen. At the present time, we are actively engaged in the production of research quantities of highly enriched oxygen-17 and oxygen-18. Most of these materials are special status products and are not listed in the current ORNL Isotopes Catalog. I shall, therefore, briefly describe the availability and cost of each of these materials.

HYDROGEN

After the development of the bubble chamber by Glaser in 1955, high-energy physicists became interested in using D_2O or liquid D_2 in these devices. Unfortunately, many experiments of this type proved impractical because of the spurious tracks created in the bubble chamber by the tritium which had been concentrated in the heavy water by the same process which was used to concentrate the deuterium. A D_2O or D_2 product depleted in tritium was needed. Utilizing a repetitive electrolytic separation process¹, the Isotope Group produced some 400 kg of D_2O having a T/D ratio of 6×10^{-16} , and about 9000 NTP liters of D_2 having the same isotopic purity. Much of this initial inventory is still available. The cost of the best grade product is about \$164 per liter. Material of lower purity is available at a proportionately lower cost.

-
1. Production of Low-Tritium Deuterium, J. S. Drury, R. H. Guymon, and E. F. Joseph. Chemical & Process Engineering, May 1961, pp 1-5.

LITHIUM

High purity lithium-6 and lithium-7 are available as the metal, Li_2CO_3 , LiH , LiF , Li_2SO_4 , LiD , and $\text{LiOH}\cdot\text{H}_2\text{O}$. The cost for 95.6% Li-6 is about \$6-\$8 per gram, depending upon the form; and 98.7-99.3% Li-6 costs \$23.00-\$25.00/g. The maximum order for this grade product is 10 grams. Kilogram quantities of 99.99% Li-7 are available as $\text{LiOH}\cdot\text{H}_2\text{O}$. The cost for this isotope is \$1.00/g or \$120.00/kg. Additional descriptions of the available materials may be found in the ORNL Catalog of Radio and Stable Isotopes.

BORON

Boron isotopes are available as crystalline and amorphous metal powders, $\text{BF}_3\cdot\text{CaF}_2$, $\text{BF}_3\cdot(\text{CH}_3)_2\text{O}$, KBF_4 , and H_3BO_3 . The isotopic purity of the boron-10 forms ranges from 92.0%-96.5%. The corresponding costs vary from \$3.50/g. to \$11.00/g. The boron-11 forms are available in isotopic purities of 89.0 to 98.4% at costs of \$10.00/g to \$13.50/g. All of the available isotopic forms of boron are in relatively short supply at the present time, and orders for 92% boron-10 metal powder are limited to 10 grams maximum. All of the above materials were separated by a chemical exchange process involving the distillation of the $\text{Me}_2\text{O}\cdot\text{BF}_3$ complex. Operation of this process was halted in 1957. A new and better method of separating boron isotopes was subsequently developed by the Isotope Group at ORNL.^{2,3} The A.E.C. is currently taking steps to augment the present limited supply of enriched boron isotopes. Further details concerning the availability of enriched boron isotopes may be found in the ORNL Catalog of Radio and Stable Isotopes.

-
2. Separation of Boron Isotopes. II. The BF_3 -Anisole System. A. A. Palko, R. M. Healy, and L. Landau. JCP 28, 214-217 (1958).
 3. Separation of Boron Isotopes in the Bench-Scale Boron Fluoride-Anisole Unit. A. A. Palko. Ind. Eng. Chem. 51, 121-124 (1959).

NITROGEN

Nitrogen-15 is now available in purities up to 95% from commercial sources. Prior to the development of commercial sources of this isotope, the A.E.C. authorized the Isotope Group to separate one kilogram of nitrogen-15 at a purity of 95% or greater. The required product was separated by means of the Nitrox Process.⁴ It is currently available in the form of a nitric acid solution in isotopic purities ranging from 90-98% nitrogen-15 at a cost of \$372 per gram of contained nitrogen-15. Because of the availability of nitrogen-15 from domestic commercial sources, distribution of the ORNL product is limited to A.E.C. sponsored investigators.

-
4. Automatic Cascade for the Production of Nitrogen-15, G. M. Begun, J. S. Drury, E. F. Joseph. *Ind. Eng. Chem.* 51, 1035 (1959).

OXYGEN

Oxygen exists in seven isotopic forms. The isotopes of mass 14, 15, 19, and 20 are radioactive, having half-lives of two minutes or less. Those of mass 16, 17, and 18 are stable; their normal abundance is 99.8%, 0.037%, and 0.20%, respectively. Until the present time, research involving oxygen tracing has been performed almost exclusively with oxygen-18. This isotope can be concentrated conveniently by several techniques, and it is currently available in purities up to ~95%. However, use of oxygen-18 is sometimes inconvenient since elaborate recovery procedures may be necessary to prepare the sample for analysis by existing methods. Some research cannot be performed at all because of conflicting conditions required in the sample recovery step. Prior to the development of the nuclear magnetic resonance (NMR) there was no alternate course; now, with the NMR spectrometer, it is feasible to use oxygen-17 as a tracer. In certain instances the analysis may even be performed without interfering with the experiment.

Use of the NMR technique depends upon the availability of oxygen enriched in oxygen-17. Small quantities of low purity oxygen-17 have recently become available, largely as a by-product of oxygen-18 enrichment, but no highly enriched material has been or is available. In response to this need, there has been constructed at the Oak Ridge National Laboratory the world's first facility designed for the production of research quantities of highly enriched oxygen-17, purities of 50% and greater. The enrichment process is a combination of two different isotopic separation methods, distillation of water, and thermal diffusion of oxygen gas. Oxygen-17 is first concentrated

from 0.037% to 3.67% in a cascade of water distillation columns. The partially enriched water from this process is electrolyzed and the resulting oxygen-17 is concentrated to 50% in a cascade of hot-wire thermal diffusion columns.

Preliminary Design Considerations

The decision to undertake the design and construction of an oxygen-17 enrichment facility was preceded by a series of preliminary studies concerning the best method to use. Chemical exchange methods having attractive separation factors for oxygen isotopes were known, but were rejected because of the expense or the uncertainties of the method of reflux. Gaseous or thermal diffusion of oxygen was found to be technically feasible but expensive. Distillation of an oxygen-containing species appeared attractive. Although this method had the disadvantage of a relatively small separation factor, it was characterized by a simple, economical mode of reflux, and offered the possibility of an inexpensive, non-corrosive process fluid. In addition, the technology was well established.

Water, oxygen and nitric oxide were considered as process materials. The use of oxygen, instead of water, involved a larger capital investment and offered few compensating advantages. Nitric oxide was potentially attractive because of the relatively large differences in the vapor pressures of its isotopic species. However, this multicomponent isotopic system was not sufficiently well characterized, at the time of this study, to permit the design of a reliable isotopic fractionation system. Thus, the equilibrium distillation of water was tentatively selected as the method of producing high purity oxygen-17.

It soon became apparent, however, that the water distillation method alone could not be used to produce 50% oxygen-17. Calculations revealed that ~100 years of uninterrupted operation would be required for such a system to achieve isotopic equilibrium. To reduce this equilibrium time to a reasonable value, it was necessary to limit the concentration of the product from the water distillation cascade to approximately 3% oxygen-17. Thus, to achieve simultaneously the objectives of high purity product and reasonable equilibrium time, it became necessary to combine the water distillation process with a second enrichment method having a favorable holdup to throughput ratio, i.e., a gas phase process. Gaseous thermal diffusion was the obvious choice for such an application. This enrichment method, though unsatisfactory as a primary fractionation process because of its low transport characteristics, was well suited as a final oxygen-17 enriching step.

Thus, our task became that of specifying the best combination of water distillation cascade and hot-wire thermal diffusion cascade which would yield 50% oxygen-17 at minimum cost. The complexity of the task is illustrated by the fact that we were dealing with a 9-component system in the water distillation cascade and a 6-component system in the thermal diffusion cascade. Furthermore, the isotopic fractionation factor, as well as equilibrium time, production rate and cost, varied strongly with operating pressure.

To cope with this situation, we used an IBM-704 computer. In the design of the water distillation cascade, the process variables were considered to be:

- a. column diameter
- b. column length
- c. size of column packing
- d. operating pressure
- e. feed rate
- f. oxygen-17 production rate
- g. oxygen-18 production rate.

We asked the computer to minimize the equilibrium time of the cascade. This was equivalent to requiring that the volume of packing be minimized or, in effect, that the cost be a minimum.

An iterative method was employed: first, reasonable values of the process variables were tentatively assigned to each column. Then, optimum values were found for the variables in the first column, holding the design of the remaining columns fixed, except that the length of the final column was permitted to vary so that the desired oxygen-18 concentration could be obtained in all cases. These optimum values were assigned to the first column and new values were computed for each remaining column. Each column was treated in turn. The procedure was repeated until no further changes were indicated.

The computer program just described provided the design characteristics for several water distillation cascades, each differing in productive capacity, cost, and equilibrium time. After detailed cost estimates were made for each cascade, one design was chosen for further development.

This cascade consisted of six 30-foot long concatenated columns, having diameters of 5.3", 2.7", 1.4", 0.5", and 0.5", respectively. The first column was designed to have 600 theoretical plates; the others, 630, 666, 915, 975, and 1002, respectively. The ratio of boil-up to product withdrawal rate was 24,800 in the first column, 6,170 in the second, 1,760 in the third, 577,188, and 164 in the three smaller columns. The design concentrations of oxygen-17 at

the bottom of each column, starting with the largest, was 0.1%, 0.27%, 0.7%, 2.1%, 3.5%, and 1.0%. The peak concentration of oxygen-17 was calculated to occur about 1/3 of a column length from the bottom of column 5. It was there that the product stream was to be withdrawn. According to the design calculations, water containing 98% oxygen-18 should be produced at the bottom of column 6.

It was now necessary to upgrade the product from the Water Distillation Cascade in a series of thermal diffusion columns. Because the water molecule had a very low thermal diffusion separation factor, molecular oxygen was chosen as the process gas.

The coefficients of viscosity and ordinary diffusivity were computed from the kinetic theory of Chapman and Enskog, using the Lennard-Jones 6-12 potential energy function to describe the potential energies of interaction.

The transport coefficients were computed from shape factors of Jones and Furry, based on their theoretical model for the extreme cylindrical case.

The effective length of an individual thermal diffusion column was set at 30 feet, and the filament temperature was fixed at 1050 °K in order to obtain rapid exchange among the several isotopic species present in the column.

A cascade of 13 interconnected columns was ultimately selected. Oxygen at the rate of 1.44 grams per day is fed from the electrolysis cell to the midpoint of five interconnected thermal diffusion columns. This gas is about 51% oxygen-18, 46% oxygen-16, and 3.7% oxygen-17. The first set of five columns fractionates this feed into two binary streams containing essentially only oxygen-16 and oxygen-17 on one hand, and only oxygen-17 and oxygen-18 on the other. The remaining columns form two secondary cascades consisting of one large and three small diameter columns each. The light fraction from the primary cascade is separated into nearly pure oxygen-16 at one end and 50% pure oxygen-17 at the other end of this (light) secondary cascade. In similar fashion, the heavy fraction from the primary cascade is separated into nearly pure oxygen-18 at one end of the other (heavy) secondary cascade, and into 50% pure oxygen-17 at the opposite end of this cascade. The design rate of production of 50% pure oxygen-17 gas is about 80 NTP cc per day.

Construction of the Oxygen-17 Separation Facility was completed last year. The Water Distillation Cascade has now been operating for 78 weeks, the Thermal Diffusion Cascade for 16 weeks. The present oxygen-17 concentration is about 1.2%, the oxygen-18 concentration about 99%.

When steady state operation is achieved, oxygen-17 will be available as H_2O the oxygen being 50% oxygen-17. The cost of this product will be \$900/gram. By special arrangement, oxygen-17 gas having an isotopic purity of 50% may be available. The cost in this form will be \$1000/gram of O_2 , plus a small conversion charge. Oxygen-18 will be available as D_2O , having an isotopic purity of 99% or greater. The cost of this material will be \$550 per gram of D_2O .

PRODUCTION OF HEAVY WATER

C. H. Ice, SRL

At the Savannah River Plant, heavy water is extracted from the Savannah River and concentrated to about ten per cent D_2O by the dual temperature H_2S/H_2O exchange process (GS Process). Further concentration, to specification quality of 99.75 mol per cent minimum is done by vacuum distillation. Capacity of this plant is about 540 tons per year; however, because of the large inventory that had been built up while both this plant and a similar plant at Dana, Indiana, were being operated, and due to the present demand for heavy water, only one-third of the Savannah River heavy water plant has been operating since late in 1958. (The Dana Plant was shut down, and is no longer available.) Demands for heavy water have been exceeding present production by well over 100 tons per year, and it is expected that an additional one-third of the Savannah River Plant will be started up within the next few years.

Tritium, which is present in small quantities in the river water, is extracted and concentrated along with the deuterium. Further, although a separate facility is provided for reconcentrating heavy water from the reactors at SRP, thus avoiding large additions of tritium to the product some heavy water containing lesser quantities of tritium is reconcentrated from time to time in the heavy water production facility. When only natural water is being fed to the production facility, the atom ratio of tritium to deuterium generally runs from 3 to 10×10^{-14} . Most of the heavy water in our stockpile contains only this amount of tritium, and effort is being made to reserve a supply of this material for use in nonreactor research. Heavy water containing larger amounts of tritium is distributed only to reactor operators.

Approximately ten tons per year of "low tritium" heavy water is currently distributed to users annually, half of this within the Commission, and the remaining to domestic customers, and universities. Heavy water is available at SRP to AEC and other government agency users at essentially the operating cost to produce the material; at the present time, this price is about \$13.60 per pound, but is expected to increase in the near future. Universities can obtain a free loan of heavy water by applying to the Commission's Division of Nuclear Education and Training if the heavy water is to be used in nuclear education. Domestic customers purchase heavy water directly from "domestic suppliers." These suppliers must specialize in the problems of repackaging in small quantities while monitoring isotopic quality. Prices from such sources therefore depend on the quantities and concentrations sold, and on the suppliers' costs and policies. Large quantities (drums of 125 lb. or more) are handled by the USAEC at the established sales price of \$24.50 per pound.

The Savannah River production facility can produce heavy water at higher D₂O concentration than 99.75 mol % if such material is needed. We have never been informed of any such need, and have not previously announced the availability of high concentration material. The cost would be about \$14 per pound for 99.9 mol % D₂O, and \$15 per pound for 99.95 mol %.

Further information regarding the availability of heavy water may be obtained by contacting C. B. Bastin of the AEC's Savannah River Operations Office.

HELIUM-3 AND OTHER GASEOUS ISOTOPES

W. J. Haubach, Mound

Mound Laboratory has a considerable number of thermal diffusion columns available, and can undertake special preparations on a research scale of other gaseous isotopes. The accompanying figures are self-explanatory.

Helium-3, enriched to 99.9996% He-3 with less than 10^{-10} per cent tritium, is available from Mound.

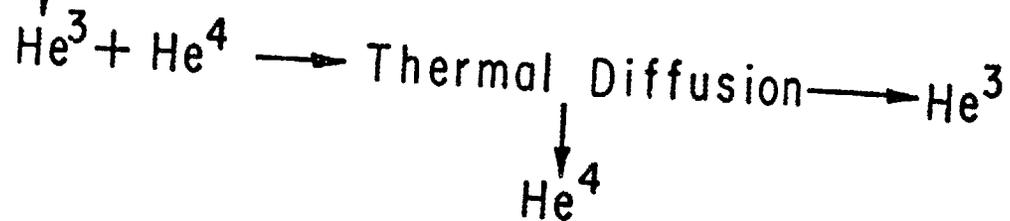
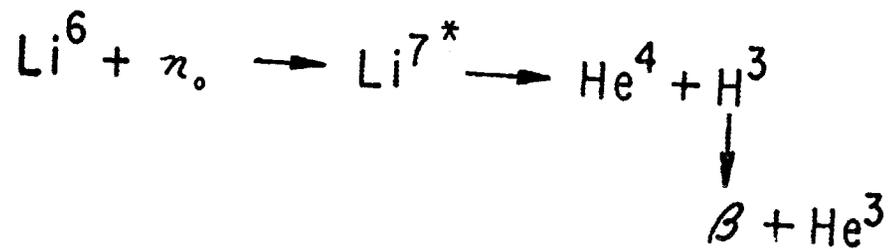


Fig. 1. Preparation and Chemical Purification of Helium.

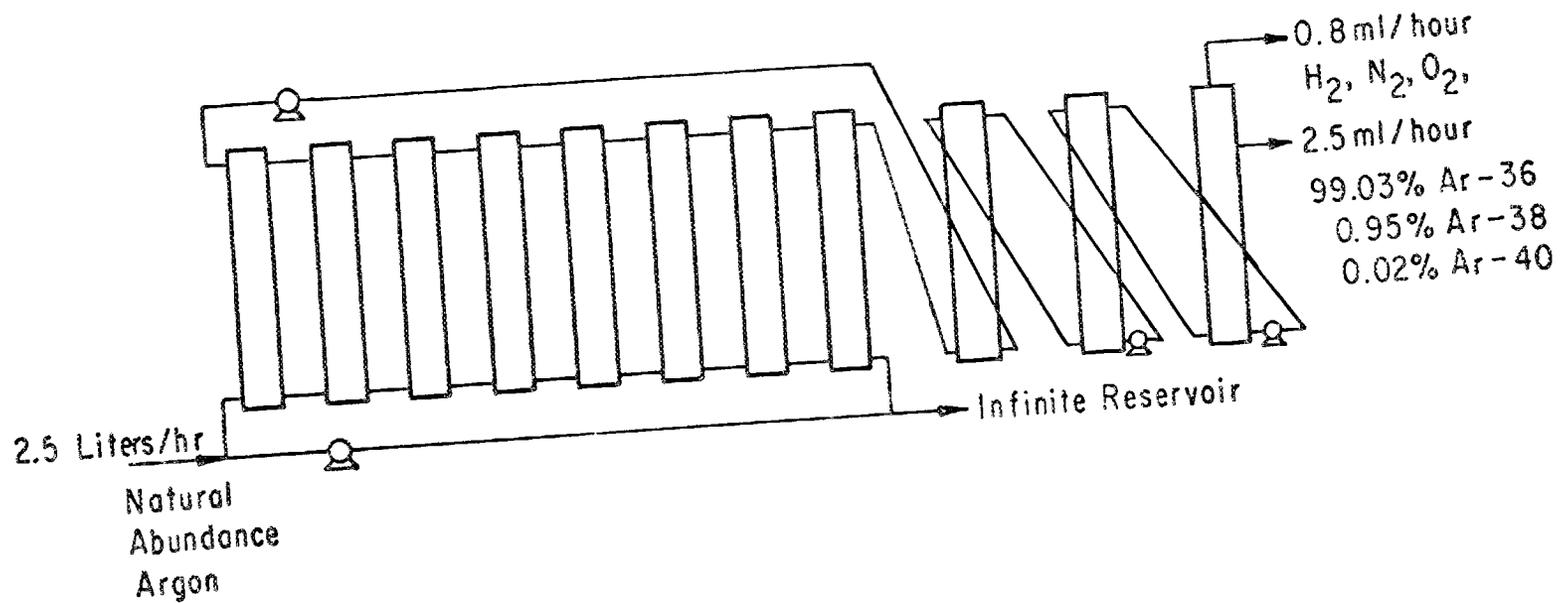


Fig. 2. Separation of Argon Isotopes by Thermal Diffusion.

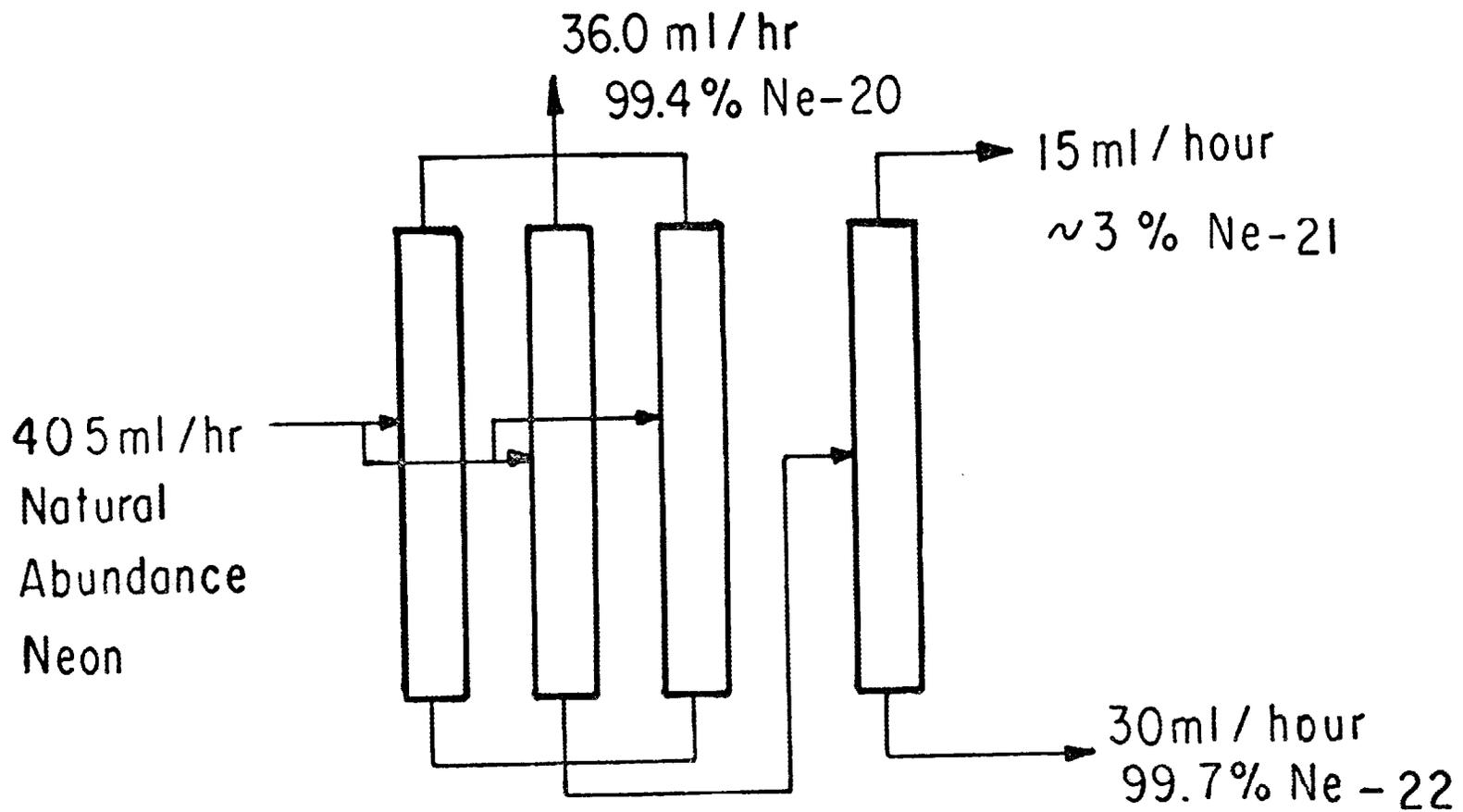


Fig. 3. Separation of Neon Isotopes by Thermal Diffusion.

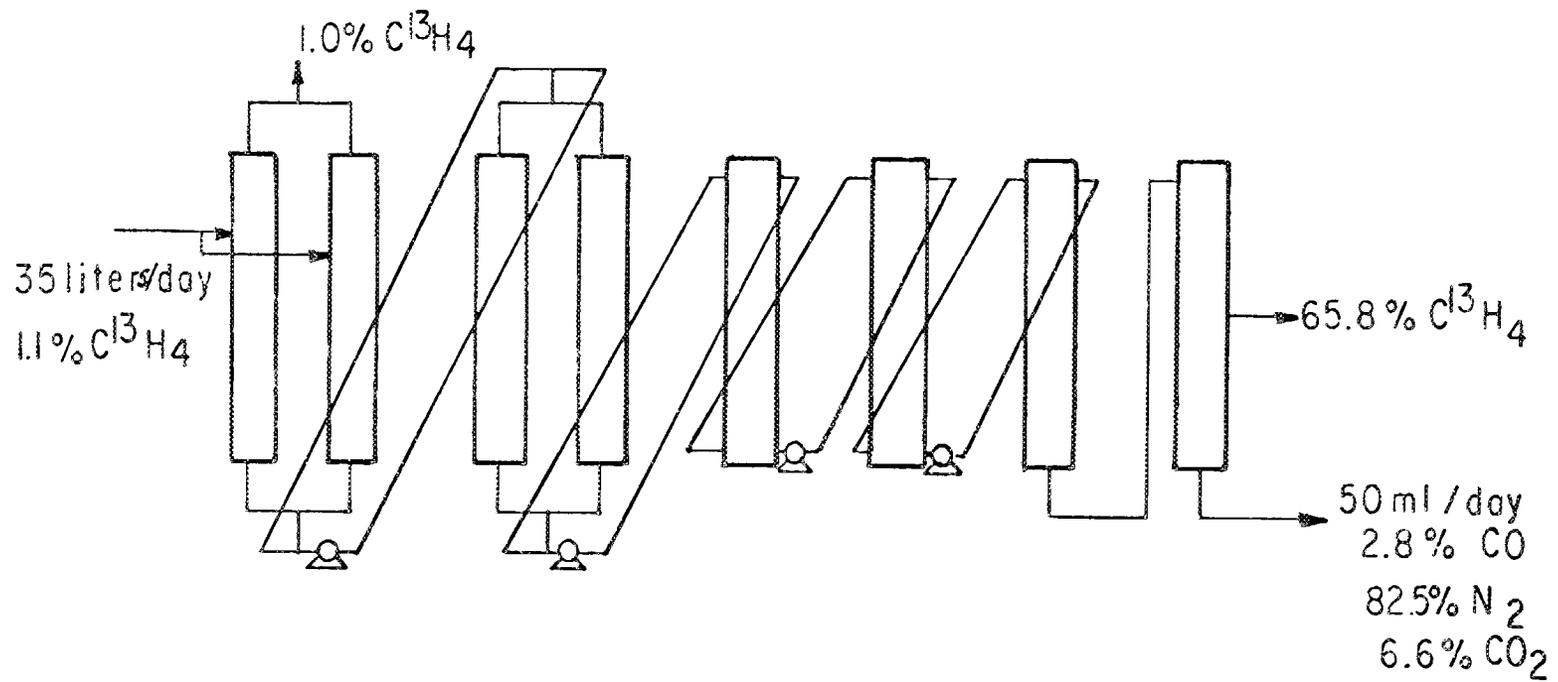


Fig. 4. Concentration of Intermediate-Level Carbon-13 by Thermal Diffusion.

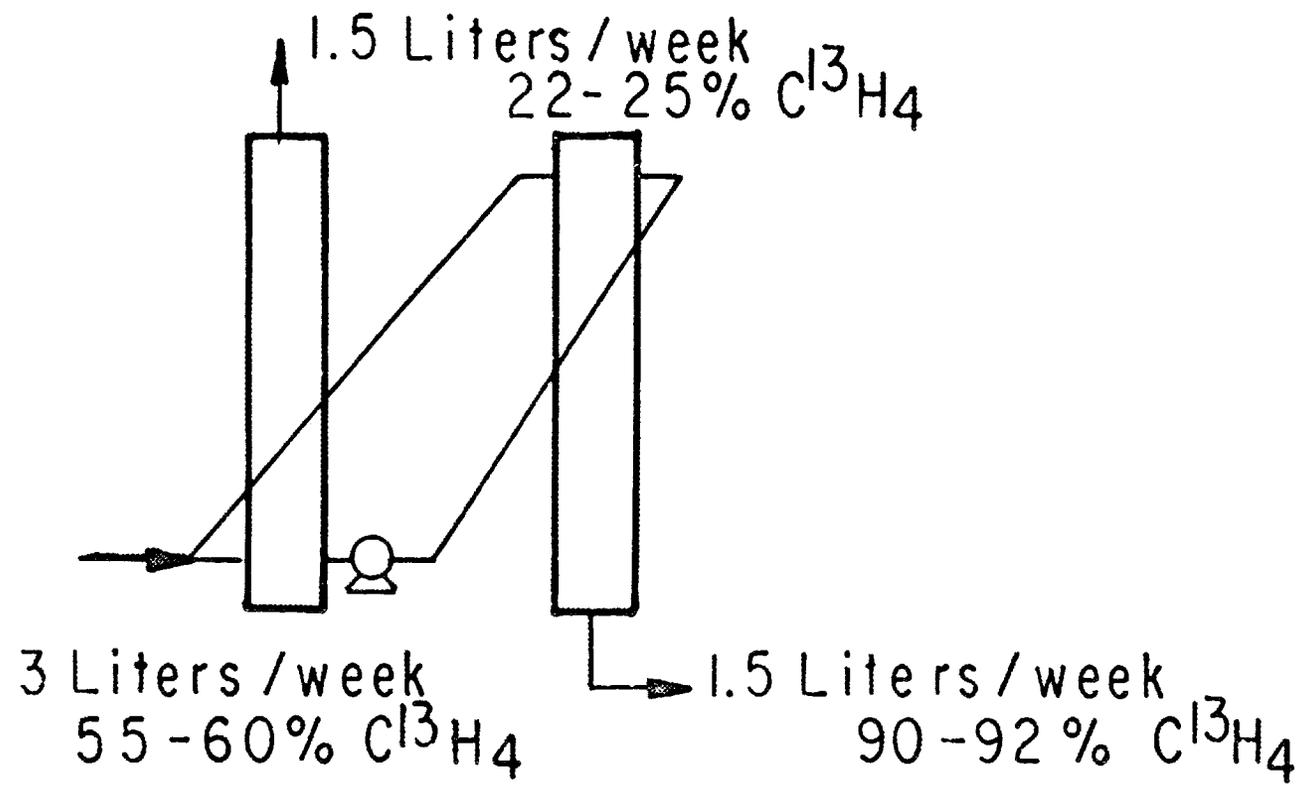


Fig. 5. Concentration of High-Level Carbon-13 by Thermal Diffusion.

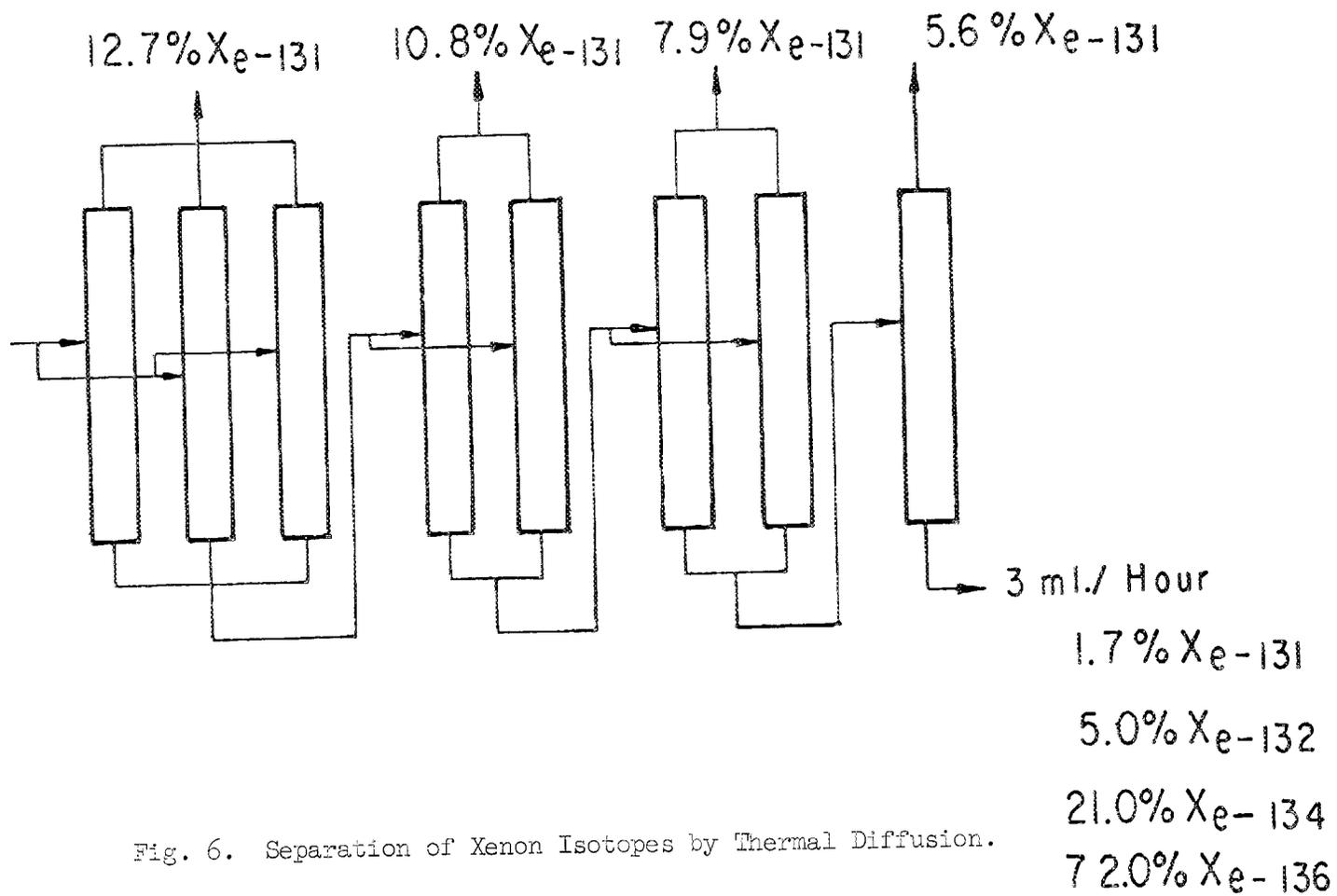


Fig. 6. Separation of Xenon Isotopes by Thermal Diffusion.

GAS INVENTORY

Gas	Isotopic Analysis	STP Liters
Helium	Classified	Classified
Neon	Natural Abundance	8,880.
	99 % 22	100.
	2.2% 21	97.
	99 % 20	1,900.
Argon	98.8% 36, 1.1% 38, 0.1% 40	5.
	96.9% 36, 3.0% 38, 0.1% 40	1.3
	24 % 36, 15 % 38, 61 % 40	1.1
	70 % 36, 15 % 38, 15 % 40	1.1
		} *
Krypton	Natural Abundance	2,030.
	1 % 78, 7 % 80, 92 % (82,83,84,86)	11.
	4.5% 78, 15.7% 80, 79.8% (82,83,84,86)	1.5
	8.5% 78, 23.1% 80, 68.4% (82,83,84,86)	1.
	0.2% 78, 1.8% 80, 98 % *82,83,84,86)	41.
Xenon	Natural Abundance	190.
	Fission	Classified
	2.7% 131, 69% 136	2.
	5 % 131, 57% 136	7.
Carbon**	10% 13 (CO)	0.3
	10% 13 (CH ₄)	0.6
	20% 13 (CH ₄)	0.2
	25% 13 (CH ₄)	2.2
	38% 13 (CH ₄)	0.4
	57% 13 (CH ₄)	3.5
	60% 13 (CH ₄)	0.4
	90+% 13 (CH ₄)	7.1

* Column Hold-up

** Quantity in Grams

Fig. 7

^{238}Pu , ^{234}U , AND NATURALLY OCCURRING RADIOISOTOPES

R. E. Vallée, Mound

The accompanying twelve slides are self-explanatory and describe the current status of the referenced isotopes. The maximum ^{234}U content which would be obtained from decay of ^{238}Pu is $> 99\%$ ^{234}U (in material which initially contained no uranium). The following table represents a typical case:

Isotope	Amount (Mg)	Per Cent
^{234}U	646	99.896
^{235}U	0.455	0.070
^{236}U	0.211	0.033
^{237}U	8.5×10^{-3}	0.001
^{238}U	9.0×10^{-5}	1.4×10^{-5}
^{233}U	1.1×10^{-4}	1.7×10^{-5}

The above calculations are based on one year's decay of a 100-g plutonium sample containing 81% ^{238}Pu , 16% ^{239}Pu , 2% ^{240}Pu , 0.5% ^{241}Pu , 0.05% ^{242}Pu , 0.4% ^{237}Np , and no uranium.

TABLE 1.

PLUTONIUM-238 FEED MATERIAL

Total Plutonium Content	$\geq 98\%$
Neptunium-237	$\leq 1\%$
All Other Cations	$\leq 1\%$
Total Fission Fragments	$< 10^{-5}$ C/gm
Pu ²³⁸ Isotopic Abundance	$80 \pm 2.5\%$

TABLE 2.
PLUTONIUM METAL - ISOTOPIC POWER

Isotope	Half-Life (years)	Average (%)	Watts/gram Plutonium
238	86.4	80.0	0.442
239	2.4×10^4	16.5	0.0003
240	6.58×10^3	2.5	0.0002
241	13.0	0.8	-
242	3.8×10^5	<u>0.08</u>	<u>-</u>
Total		100.	0.442

TABLE 3.

TYPICAL NON-RADIOACTIVE IMPURITIES (%)

Boron	0.01	Aluminum	0.06
Magnesium	0.004	Copper	0.008
Manganese	0.02	Sodium	0.11
Chromium	0.004	Titanium	0.008
Iron	0.14	Calcium	0.06
Nickel	0.06	Silicon	0.004

0.50% Total Impurity

PREPARATION OF PLUTONIUM-238 METAL

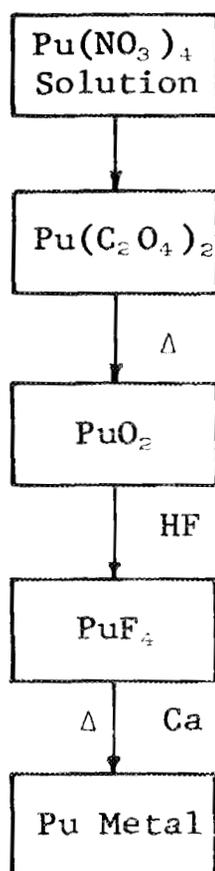


Fig. 1

TABLE 4.

ANALYSIS OF PLUTONIUM METAL

<u>Elements</u>	<u>Analytical Method</u>	<u>Analysis in Weight Per Cent</u>
Pu (Total)	Chemical	99.13
Cd	Emission spec.	> 0.15
Al	Emission spec.	0.15
Na	Emission spec.	trace
Ca	Emission spec.	trace
Zn	Emission spec.	N.D.
Fe	Colorimetric	0.15
Mg	Emission spec.	trace
Pb	Emission spec.	trace
B	Emission spec.	0.09
Ni	Emission spec.	0.05
Bi	Emission spec.	N.D.
Cr	Emission spec.	0.05
Mn	Emission spec.	0.03
Cu	Emission spec.	0.03
Ti	Emission spec.	trace
Pu ²³⁸	Chemical	78.43 of Total Pu
	Mass spec.	78.62 of Total Pu
	Alpha Counting	77.76 of Total Sample

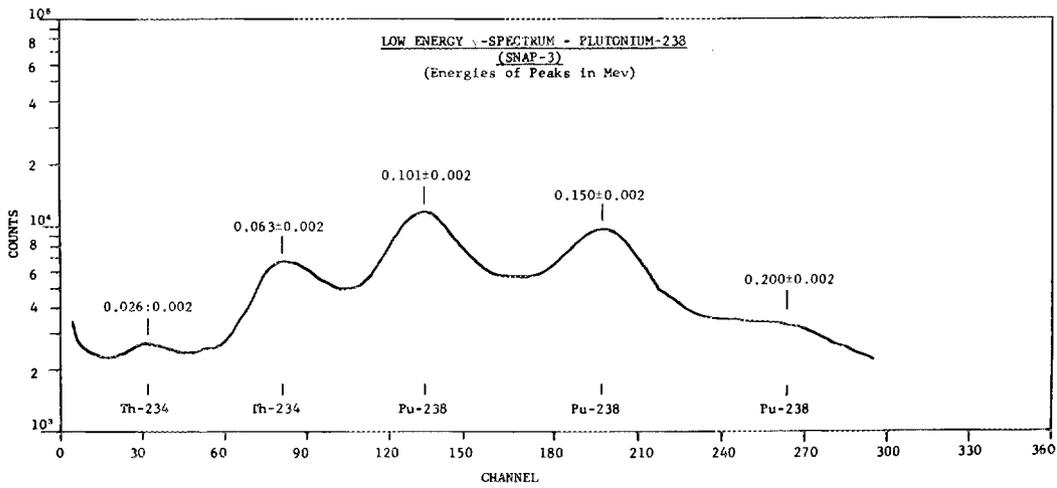
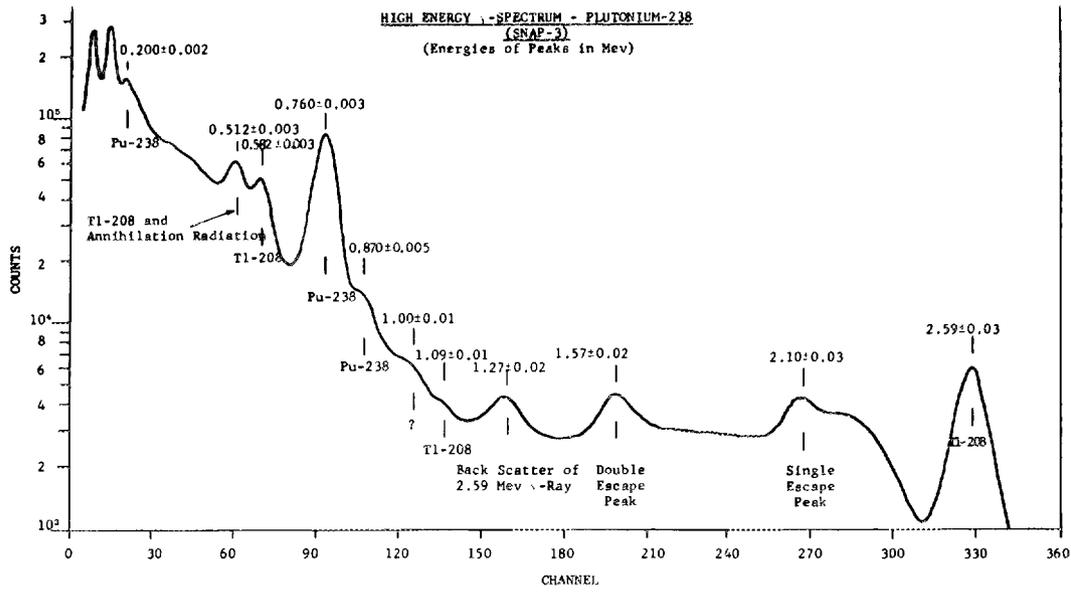


Fig. 2

TABLE 5.

Pu-238 Radiation

	γ (mr/hr/gm @ 1 meter)	n° (n/sec/gm)
Metal	4×10^{-3}	3×10^3
Oxide	4×10^{-3}	2×10^4

TABLE 6.

QUANTITIES OF U-234 READILY AVAILABLE

Lot	Quantity (mg)	Status
1	~ 60	Processed
2	~ 200	In Process
3	~ 250	} Available for Processing
4	~ 325	
5 (SNAP III)	~ 1800	
6 (Oxide Cow)	~ 380	

URANIUM-234 RECOVERY PROCESS

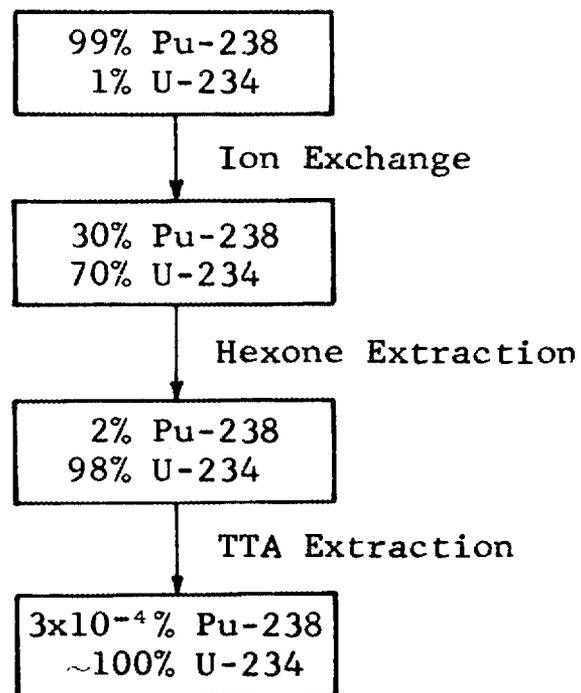


Fig. 3

TABLE 7.

TYPICAL U-234 ANALYSIS

U-234	98.0 %
U-235	0.33 %
U-236	0.15 %
U-238	1.54 %
U-232	n.d. by mass spec.
Pu-238	$\sim 3 \times 10^{-4}$ %

SEPARATION AND PURIFICATION OF ACTINIUM-227

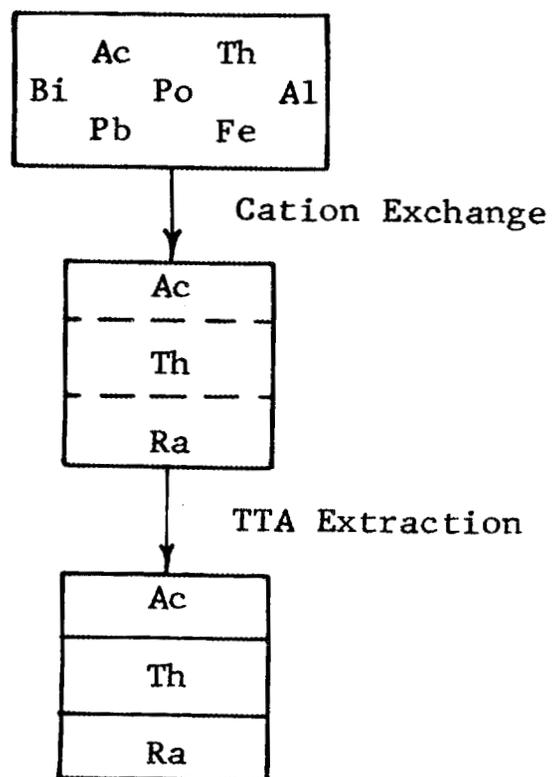


Fig. 4

TABLE 8.

PLANNED Ac-227 AVAILABILITY

Form	Amounts
Nitrate Sol'n	μ curie to m curie
Chloride Sol'n	μ curie to m curie
Oxide	curie

CHEMICAL AND SPECIAL PROCESSING OF ELECTROMAGNETICALLY SEPARATED ISOTOPES

H. R. Gwinn, ORNL

By way of introduction I'd like to point out that we have a group of about 15 chemists working with calutron-separated isotopes who at one time or another have worked with practically every naturally occurring stable and radioactive isotope in the periodic table. These men are not primarily concerned with pushing back the frontiers of analytical chemistry but are vitally interested in improving recovery, refinement, and handling techniques for enriched isotopes. The work is divided into 2 broad categories: radioactive and stable isotopes. An interesting example of the type of special processing, which is in addition to the normal chemical recovery and refinement of calutron-separated isotopes, is illustrated by the ^{233}U ETA Program. Several years ago a calutron separation of ^{233}U was performed to provide a very high-purity material with which to make precise ETA measurements. About 12 g of $> 99.999\%$ isotopic purity was obtained from the separation. This material was then chemically refined to a high degree, converted to UF_4 , and reduced to metal suitable for rolling into foil.

In the near future, we plan to make another calutron separation of ^{233}U for use in studying the energy dependence of the fission to capture ratio. The object of this separation is not so much to obtain high isotopic purity ^{233}U as it is to reduce the concentration of the unwanted ^{232}U isotope. About 220 g of ^{233}U feed with a ^{232}U content of 0.6 ppm is available for this separation. We plan to collect about 50 g with the ^{232}U concentration reduced to the order of 5-10 parts per billion (ppb).

Along somewhat the same line, but for an entirely different purpose, ~ 3 kg of high-purity ^{238}U was separated in the calutron and placed in the Research Pool. Approximately 1,700 g of this material was in the range of 5-20 ppm ^{235}U . Although no specific requests for high-purity ^{238}U were known at that time, the inventory was quickly depleted by those whose experiments were apparently just waiting for material of this quality.

In addition to recovery, refinement, and compound conversion, the group has prepared foils or sources from enriched uranium isotopes, ^{237}Np , and ^{241}Am by electrodeposition. Some of these sources have been used as

alpha calibration standards, while very thin deposits of ^{241}Am have been used to check gamma resolution. Still other electroplated deposits of ^{235}U and ^{237}Np have been used in the fabrication of fission chambers.

The fact that the calutron is a unique tool is illustrated by its inherent quality of not only providing enriched isotopes but also performing chemical separations. Several years ago when the separation of calcium was undertaken to obtain milligram quantities of ^{46}Ca for medical research, large amounts of ^{40}Ca were collected as a by-product. Since the natural abundance of ^{40}Ca is nearly 97%, there was very little demand for the enriched isotope. Accordingly, there was little impetus for purifying the ^{40}Ca . However, one batch of 40-50 g was refined and analyzed for impurities by emission spectroscopy. The ^{40}Ca was found to be so pure with respect to Sr that it was used for spectrographic standards. As you can see, it was quite easy for the calutron to separate the Sr isotopes at mass positions 84-88 from ^{40}Ca , while it is rather difficult to separate these elements by chemical means.

In connection with low-level impurities, our chemists in stable isotope processing recently encountered an extremely difficult contamination problem. About 13 g of ^{48}Ca enriched to > 95% was used to prepare a single crystal of CaF_2 for studying the double-beta-decay of ^{48}Ca . After the crystal was put into service, a significant amount of alpha activity was found to be originating from within the crystal. The level of activity was found to be about 94,000 counts per day as compared with 3,000 counts per day for normal material. You can imagine the concern when you realize that 13 g of ^{48}Ca represented the combined output of 8 calutrons operating 24 hr a day for ~ 18 months and that many months of development effort were required to produce a single crystal of CaF_2 . Since our calutron operations are located in the Y-12 Plant Area, it was naturally assumed that the unwanted alpha activity was due to uranium contamination. Based on that assumption, the amount of uranium corresponding to the activity level was estimated to be < 1 ppm or ~ 10 μg total in the entire crystal. Usually one considers impurities at the ppm level indicative of high-quality chemical refinement. Nevertheless, the alpha activity completely eliminated the usefulness of the crystal and we were requested to repurify it. With some misgivings we agreed to attempt the repurification. There

was some doubt that we could reduce the uranium content; in fact, working in a uranium environment might indeed even increase the level of contamination. Further, we were not aware of a suitable method of analysis that could measure uranium in calcium at these low levels. During a discussion of the analytical problem with Dr. Cameron of the ORNL Mass Spectrometry Laboratory, he advised us of an isotope dilution technique which could measure uranium accurately to the ppb level. When the crystal was returned, it was crushed and dissolved in perchloric acid and sampled for uranium analysis. The result of this analysis indicated the uranium contamination level to be 27.5 ppb rather than the estimated 1,000 ppb. These results were received with mixed feelings -- glad, on one hand, that our normal refinement procedure had reduced the uranium contamination to such a low level; distressed, on the other hand, because at this low level it seemed impossible to reduce contamination much further. However, after careful repurification, the uranium content was reduced to 0.5 ppb and the sample made ready for refabrication into a single crystal.

Shown in the next slide (Figure 1) are two metallic cylinders of calutron-enriched tungsten-184. These cylinders weigh approximately 75 g each and were prepared by pressing finely divided tungsten metal and sintering at a high temperature. The metal was reduced from WO_3 with hydrogen at a controlled temperature to provide an optimum particle size for pressing. An organic binder was blended with the metal powder and the mixture was pressed into a cylindrical shape. The pressed forms were sintered at $2200^\circ C$ under a hydrogen atmosphere and were about 90% of theoretical density.

Pictured in the next slide (Figure 2) are samples of high-purity titanium and zirconium crystal bars prepared by the van Arkel process. This process, in which the relatively pure metal powder reacts with iodine and the compound is subsequently decomposed by the hot filament to form high-purity metal, is the only known method for preparing malleable forms of these elements. Excellent target foils have been rolled from many isotopically enriched metal samples prepared by this technique.

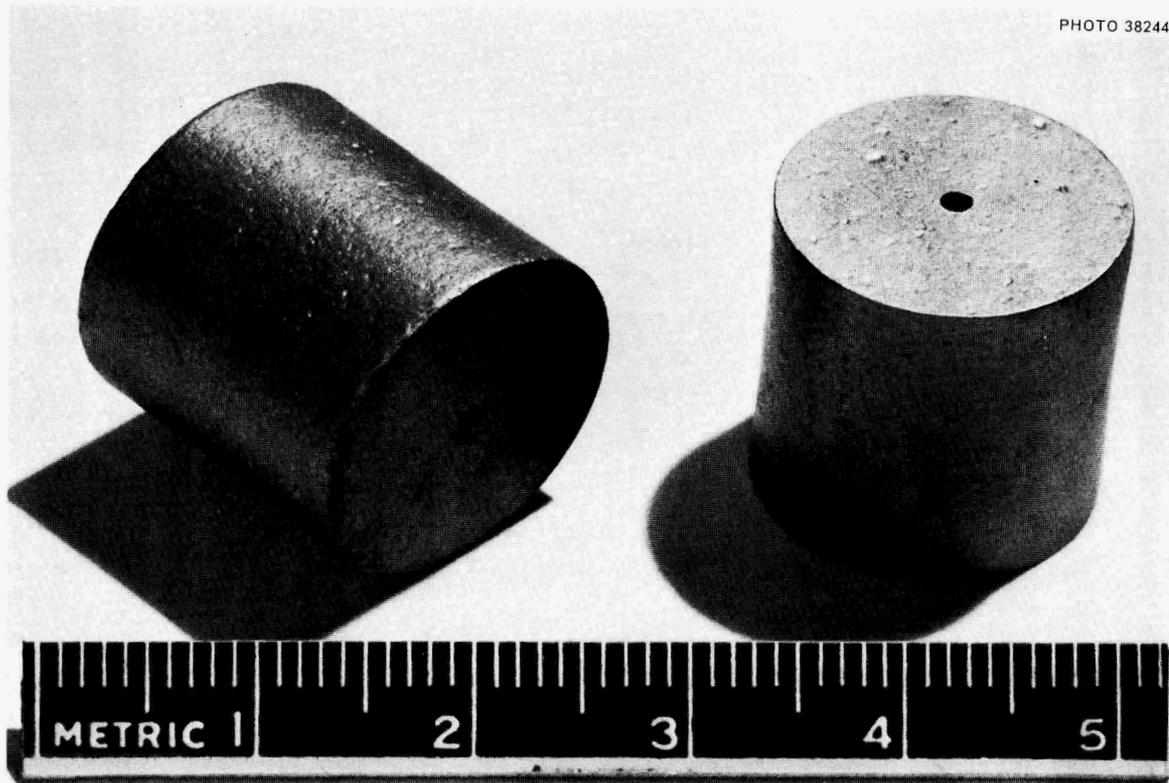


Fig. 1. Metallic Cylinders of Calutron-Enriched Tungsten-184.

PHOTO 37922

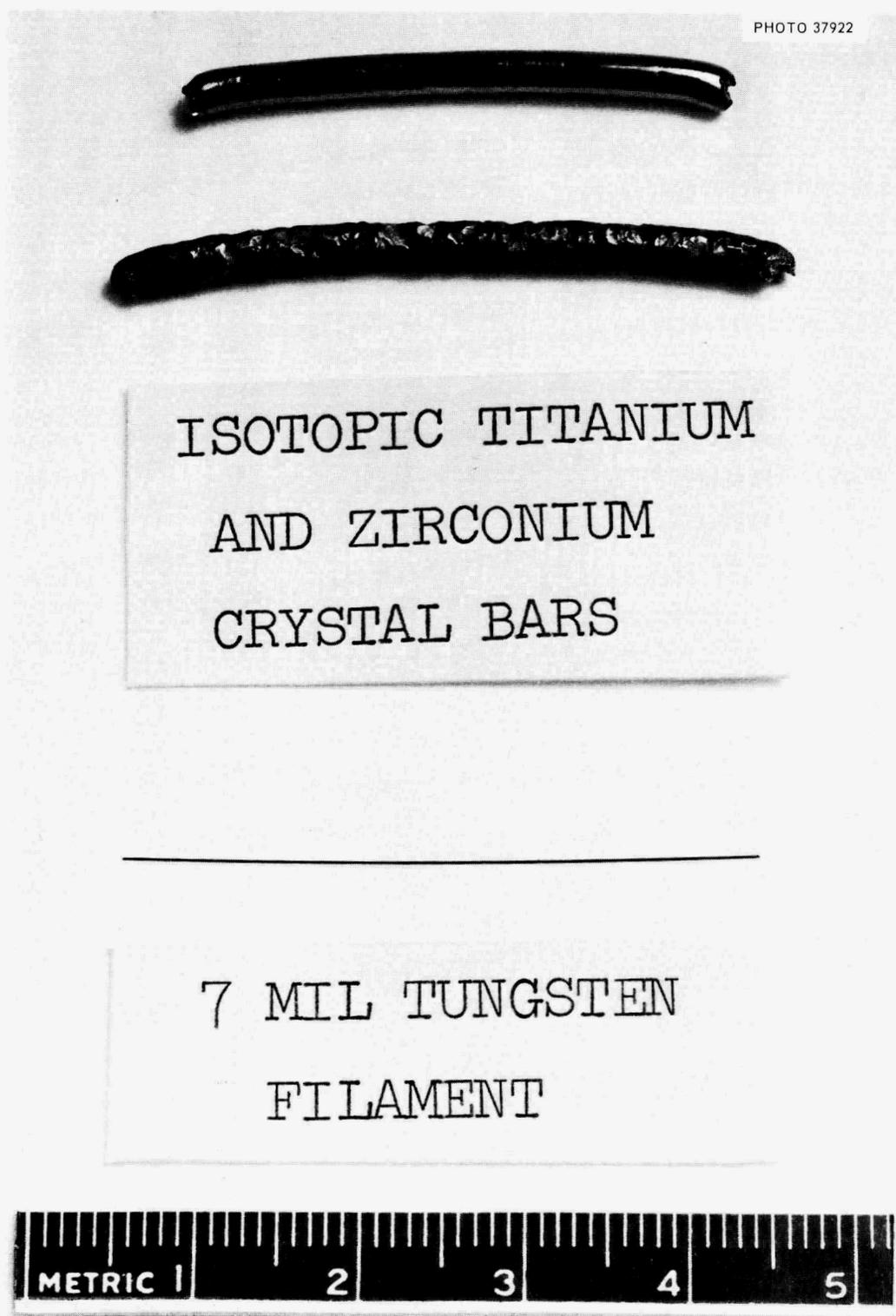


Fig. 2. Samples of High-Purity Titanium and Zirconium Crystal Bars Prepared by the van Arkel Process.

TRANSPLUTONIUM ELEMENTS - PRESENT AVAILABILITY

J. S. Lichliter, LRL

As you know, the availability of these isotopes can change very rapidly, thus anything I say now may be changed in the near future. I shall confine most of my remarks to the status of only two elements at this time, namely berkelium and californium, because these are the major concern of most researchers.

Recently ~ 5.7 μg of ^{252}Cf was transferred to Oak Ridge National Laboratory (A. Chetham-Strode) for AEC distribution purposes. Prior to this transfer of material, most authorized requests for ^{252}Cf were furnished from Lawrence Radiation Laboratory, Berkeley.

In addition to the transfer of ^{252}Cf mentioned, LRL has sent Cf to the following laboratories:

- 1.8 μg to Argonne (Ruth Sjoblom)
- 1.6 μg to Los Alamos Scientific Laboratory (Mr. Smith)
- 1.0 μg to Brookhaven National Laboratory
- 3.0 μg to Oak Ridge National Laboratory (Baybarz)
- 0.3 μg to Oak Ridge National Laboratory (Chem. Tech. Div.)

Lawrence Radiation Laboratory has on hand ~ 50 μg of Cf being used in various programs, target work, fission studies, etc. Some came from old LRL irradiated materials.

The cooperative materials in Idaho reactors are estimated as follows:

	<u>Est. Cf content</u> <u>(January 1964)</u>
UCRL-29-1 (LRL plans to process this about January 1963)	40 μg
UCRL-29-2 (Plans unknown - Don Stewart at Argonne may have more recent information)	40 μg
UCRL-29-3 (Present plans are that this may be processed on a joint venture at Mol, Belgium)	30 μg
UCRL-29-4 Processed and distributed as indicated above	
UCRL-1B-154 (113 mg ^{244}Cm) inserted in reactor September 30, 1963	
UCRL-1B-151 (850 mg ^{244}Cm) of ~ 40 μg February 1964, or 90 μg August 1964 (estimated)	
UCRL-1B-150 (100 mg ^{244}Cm) in reactor March 29, 1963; may be processed about May 1964	~ 25 μg

Berkelium-249 is in very short supply at this time. Additional information may be known following the various processing programs over the next 6 to 9 months.

Lawrence Radiation Laboratory is always interested in the various research programs concerning the transplutonium elements and welcomes the opportunity to discuss this area with representatives from the other laboratories.

TRANSURANIUM ISOTOPES
NATIONAL TRANSURANIUM PROGRAM

A. Chetham-Strode, ORNL

A number of transuranium isotopes have been or will be produced under the National Transuranium Program sponsored by the AEC's Division of Research. This program includes the interim irradiations of ^{239}Pu , ^{242}Pu , and ^{241}Am , the irradiation of kilogram quantities of ^{239}Pu to produce High Flux Isotope Reactor (HFIR) targets of ^{242}Pu , ^{243}Am , and ^{244}Cm , and the HFIR irradiation of such targets to produce gram quantities of berkelium and californium and smaller amounts of heavier transuranium isotopes and other transuranium isotopes of special interest. The present status and tentative schedule for these irradiations (revised to June 1964) are summarized in Table 1. The isotopes listed in the table (together with their separated daughters and re-irradiation products) are being produced for research activities; their distribution, foreign and domestic, is limited to small quantities for this purpose.

Requests for sources, samples, or compounds of these transuranium isotopes should be made to the U. S. Atomic Energy Commission, Division of Research.

Table 1. National Transuranium Program Irradiations

<u>Target Isotope</u>	<u>Amount</u>	<u>Product</u>	<u>Yield and Date</u>	<u>Status</u>	<u>Notes</u>
Am ²⁴¹	7 gms	Bk ²⁴⁹	~ 20 μ gm (Aug. 1964)	In pile	"Interim" irradiation
		Cf ²⁵²	~ 70 μ gm (Aug. 1964)		
Pu ²³⁹	Kilograms	Pu ²⁴²	450 gms	Processed	Target materials
			~ 500 gms	In pile	For HFIR
		Am ²⁴³	150 gms	In pile	
			~ 200 gms		
		Cm ²⁴⁴	150 gms		
	~ 200 gms	In pile			
Pu ²⁴²	60 gms	Bk ²⁴⁹	.06 mgm. (Jan. 1966)	In pile	HFIR Target Test
		Cf ²⁵²	.2 mgm. (Jan. 1966)	In pile	Elements
Pu ²⁴²	Several hundred	Bk ²⁴⁹	0.3 mgm. (May 1966)	Planned	HFIR Criticality
Am ²⁴³	Grams Initial HFIR		7 mgm. (Nov. 1966)	Planned	Scheduled May 1965
Cm ²⁴⁴	Loading	Cf ²⁵²	1 mgm. (May 1966)	Planned	
			70 mgm. (Nov. 1966)	Planned	
		Cf ²⁴⁹	mgms. (Feb. 1967)	Planned	
		Cm ²⁴⁸	mgms. (Feb. 1967)	Planned	
		Others	Starting (Feb. 1967)	Planned	

THE OAK RIDGE NATIONAL LABORATORY CURIUM PROGRAM

E. Lamb, ORNL

Oak Ridge National Laboratory is responsible for providing capability for the recovery and purification of ^{242}Cm and ^{244}Cm , the fabrication of isotopic power sources containing curium, and their characterization with respect to normal operational and extreme conditions. Curium-242 power sources will be used with thermoelectric converters to provide power for prototype testing, leading to the use of such units on space vehicles for specific missions, such as SNAP-11 or Surveyor. Curium-242 will also be used to test prototype thermionic converters (SNAP-13 Program).

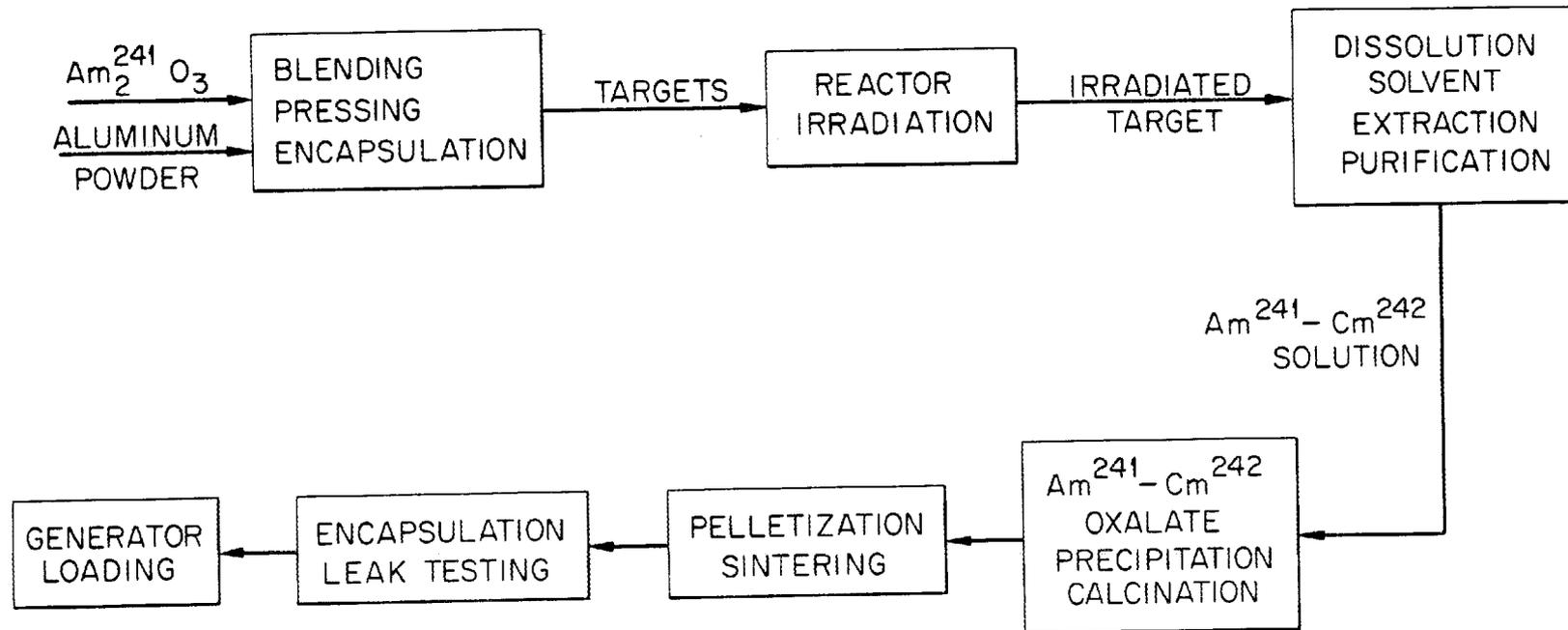
The operations involved in the preparation of ^{242}Cm heat sources (Fig. 1) are ^{241}Am target fabrication, reactor irradiation, recovery and purification of ^{242}Cm - ^{241}Am from irradiated targets, and preparation of the source form.

A target capsule (Fig. 2) contains americium as the oxide mixed with aluminum powder as a means to increase the thermal conductivity of the pellet and prevent melting of the inner portion during reactor irradiation. Up to 18 capsules will be contained in an irradiation assembly (Fig. 3) which can be used with slight modifications in several pool-type reactors. Potential uses of ^{242}Cm may require the irradiation of several hundred grams of ^{241}Am annually.

Specifications for ^{242}Cm used in SNAP units require that the gamma-emitting fission products formed during irradiation be reduced to an extremely low value. The necessary high degree of separation from fission products will be accomplished by the Tramex Process (Fig. 4), which was developed at ORNL for use in the Transuranium Program.¹ The separation will be effected in mixer settler units installed in a heavily shielded manipulator cell.

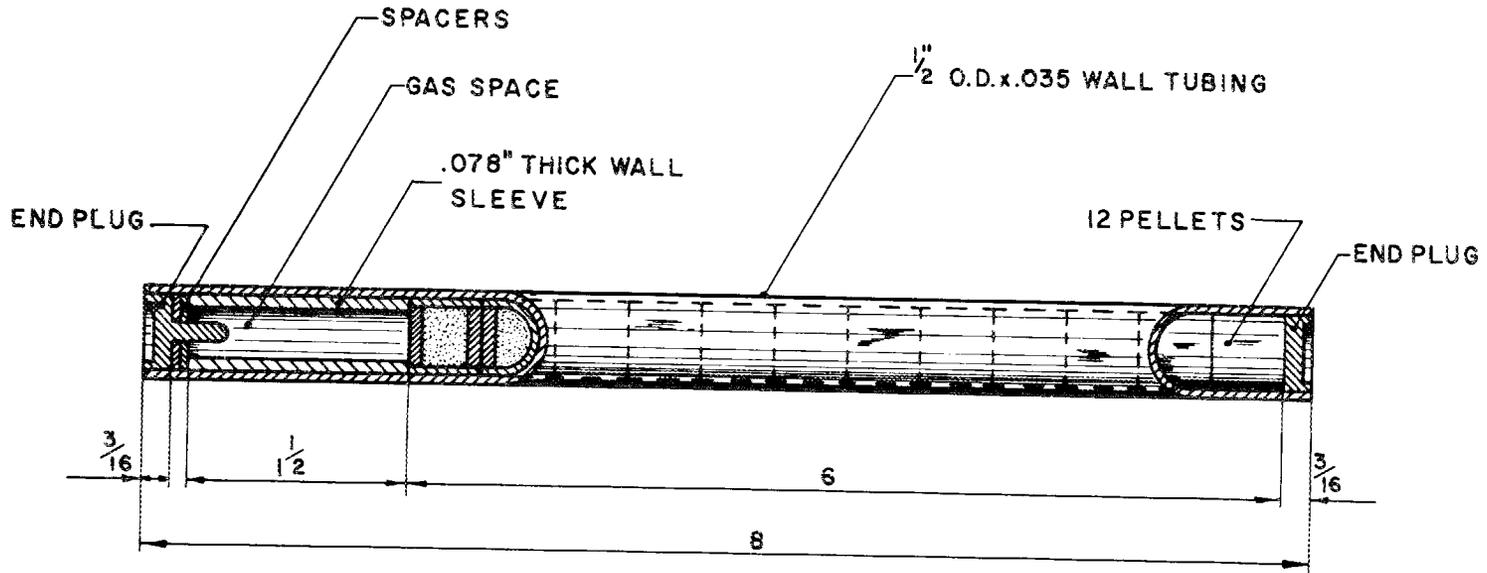
The fabrication of curium sources will be accomplished in the Source Fabrication Facility (Fig. 5), which consists of four water-shielded manipulator cells and one concrete-shielded cell. The operations performed in the SFF are shown in Fig. 6.

¹R. D. Baybarz et al., Isolation of Transplutonium Elements by Tertiary Amine Extraction, Nucl. Sci. and Eng., 17, 457-62 (1963).



Flowsheet; Preparation of Curium-242 Heat Sources.

Fig. 1



TARGET CAPSULE ASSEMBLY
ALL MATERIAL -- 1100 ALUMINUM

Fig. 2

PHOTO 60386

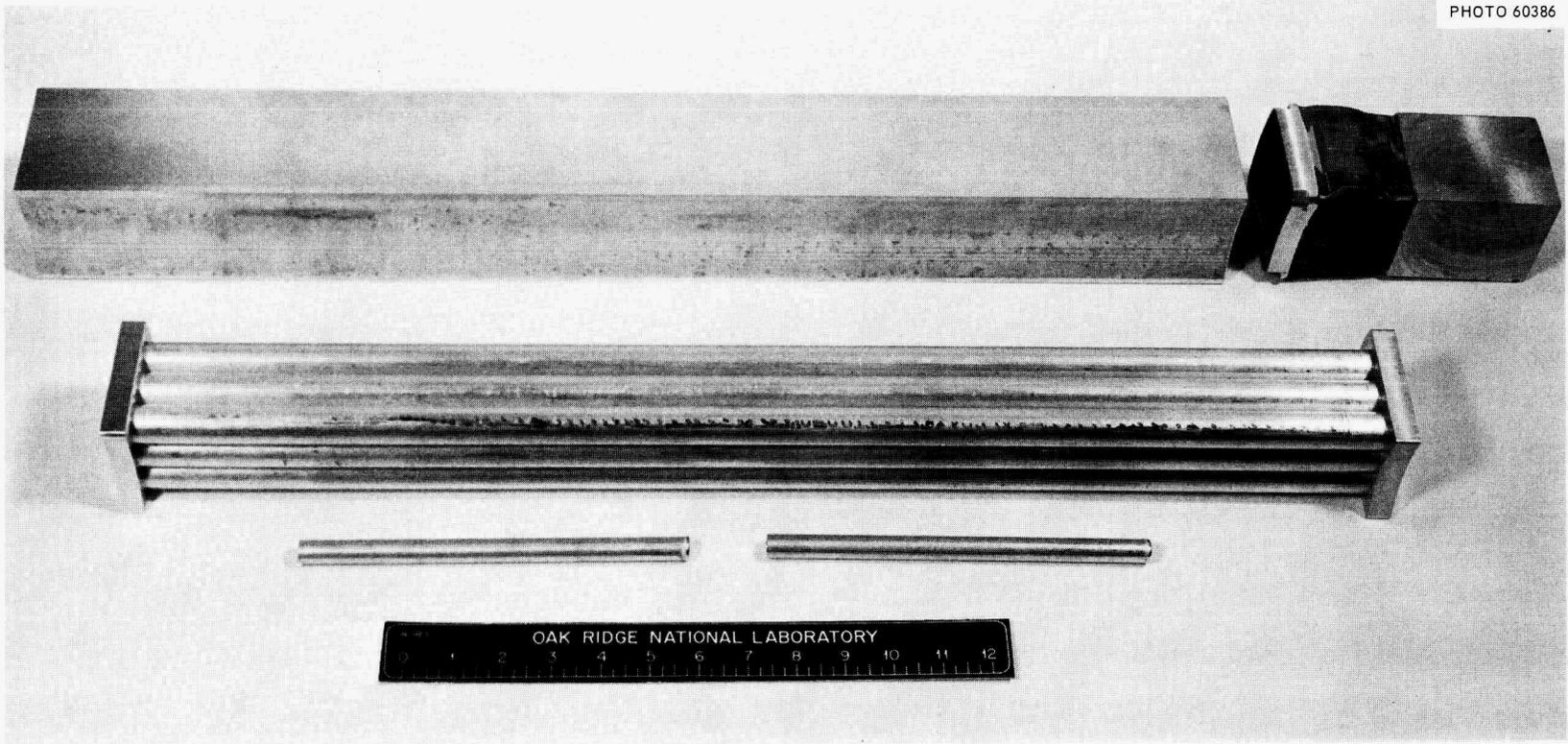
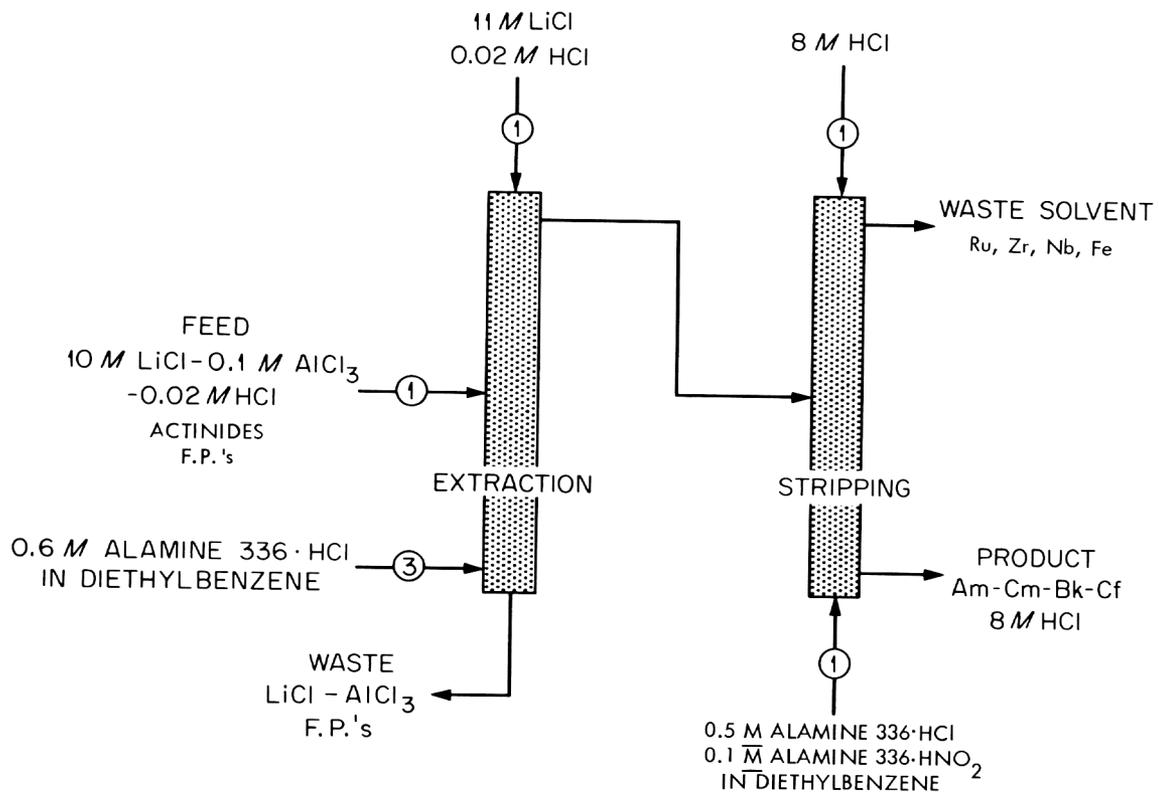


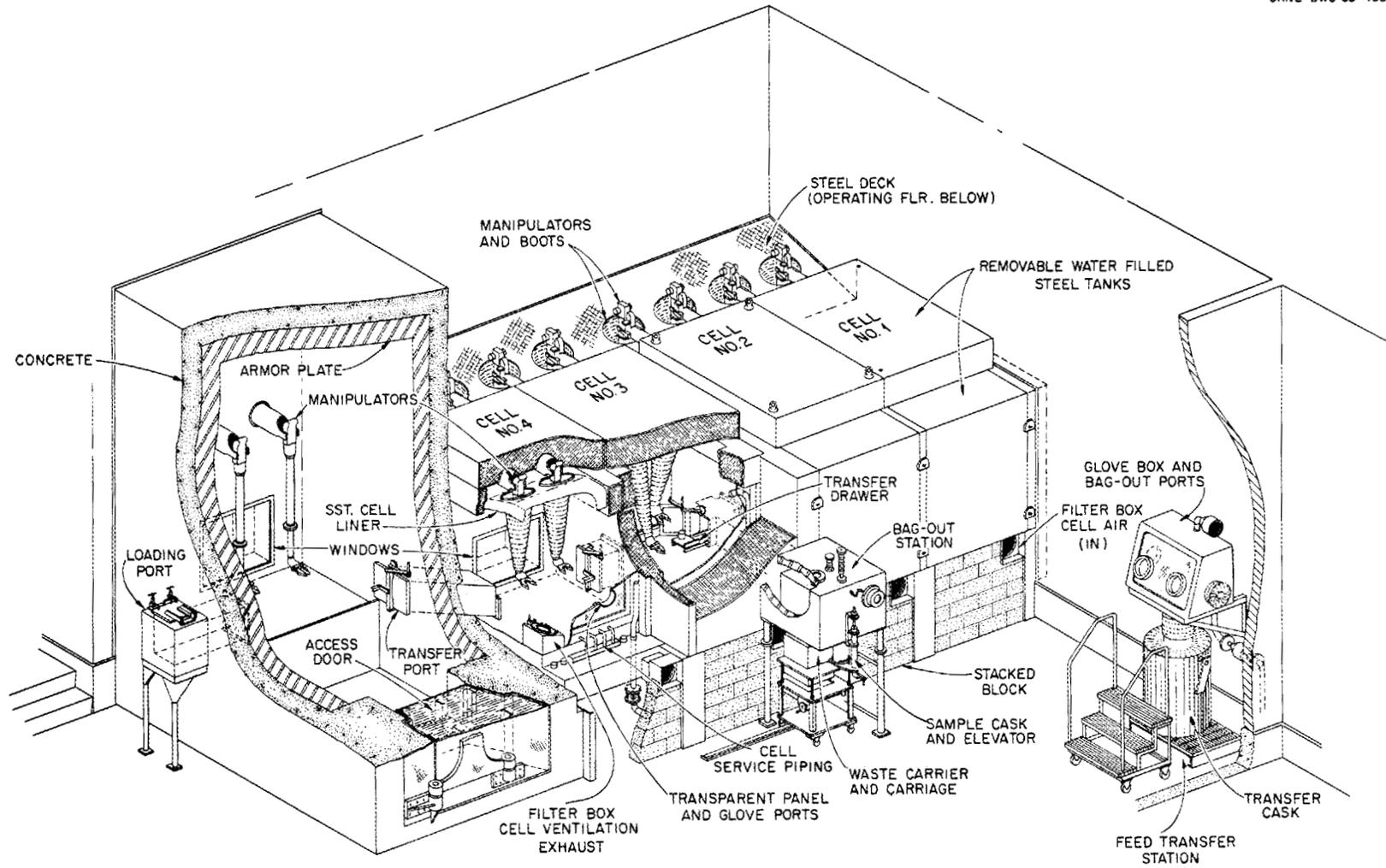
Fig. 3. Irradiation Assembly.

ORNL-DWG 72330A



TRAMEX PROCESS FLOWSHEET

Fig. 4



67

Fig. 5. Source Fabrication Facility.

CURIUM-242 POWER SOURCE PROGRAM
SOURCE FABRICATION OPERATIONS

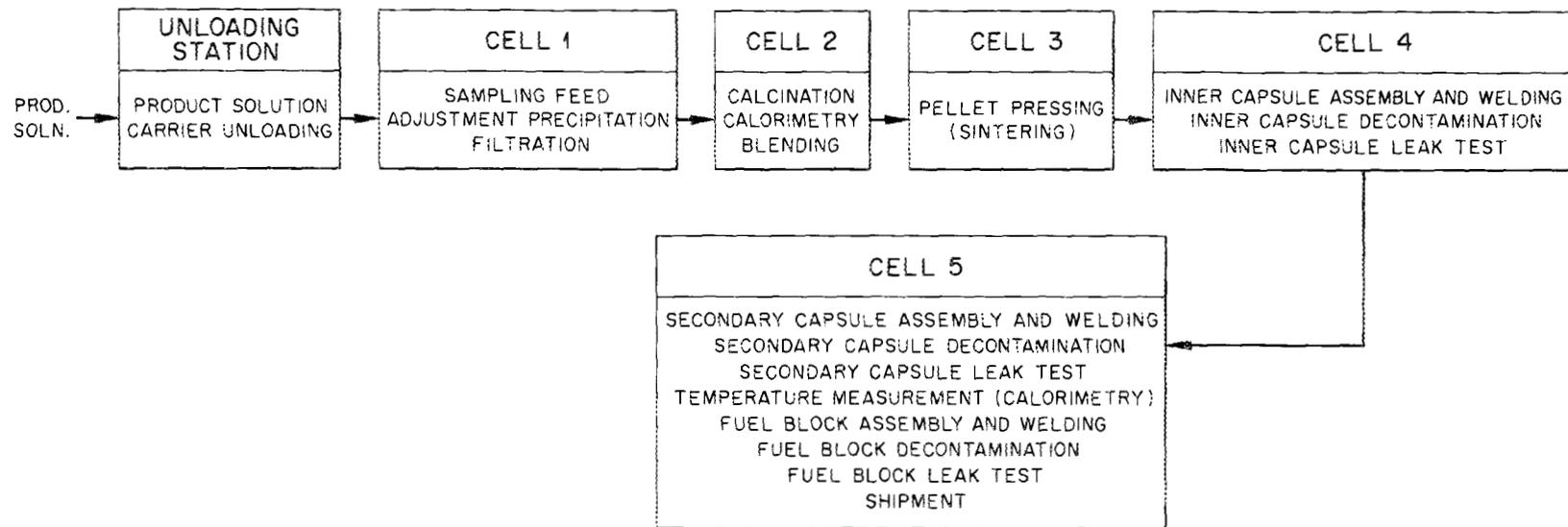


Fig. 6

PROTACTINIUM-231 AND URANIUM-232

D. E. Ferguson, ORNL

Protactinium-231

Protactinium-231 occurs as a decay product of uranium-235 and is therefore found in uranium-bearing ores. The concentration of protactinium in a uranium ore is comparable to that of radium, but because of its erratic chemical behavior is much more difficult to recover from the ore. In most cases, it is only practical to recover protactinium from very rich uranium ores such as pitchblende.

In the past, small quantities of protactinium-231 were recovered and isolated in the United States and Germany. The largest amount, about 1 gram, was obtained by Mound Laboratory from uranium processing tailing. The present major Western world supply was obtained by the British. They had thoughtfully kept the raffinates and sludges from their pitchblende processing separated from others. We in the US were not so foresighted and mixed our meager amounts of pitchblende with other ores, greatly diluting the protactinium that they contained. About three years ago, the British reprocessed their raffinate and sludges for uranium recovery and they included steps to also recover and concentrate the protactinium. In this way, they were able to obtain about 150 grams of Pa-231. The US was able to obtain about 25 grams of this protactinium in exchange for U-233, and this 25 grams represents the total available for research in the US today. Of the 25 grams, 17 has been allocated for research uses, leaving about 8 grams in the pool today. This may be obtained from the research pool in the same manner as any other special research material.

Protactinium-231 is the most stable isotope of protactinium with a half-life of 32,480 years. The next most stable isotope probably is Pa-233 with a half-life of 27.4 days. Therefore, Pa-231 is in demand for doing macro-chemical research on compounds and solutions of protactinium. In addition to this, the physicists have been most interested in adding the nuclear constant of this isotope to their charts.

It should be added that it is very difficult to accurately analyze small quantities of Pa-231. For example, with a reasonable amount of effort, a solution sample containing protactinium can be analyzed to at best $\pm 10\%$ by alpha pulse height analysis. For more accurate methods, one must resort to gravimetric analyses.

It should also be added that while we have put a price on protactinium, the present supply cannot be replaced for any reasonable amount of money, so it should be conserved as a precious research material.

Uranium-232

The isotope uranium-232 does not occur in nature. We here at ORNL have prepared two samples of U-232 by the neutron irradiation of 50 grams of protactinium-231 loaned by the British for this purpose. The production of U-232 involves 1 neutron capture and a β decay; however, U-233 is also made by the capture of an additional neutron in U-232 or the intermediate Pa-232.

The first sample prepared was made by a 7-hour irradiation of 50 grams of protactinium-231. This produced 32 milligrams of U-232 containing only 160 parts per million of U-233 plus 100 parts per million of U-235 from the natural uranium contamination of Pa-231 sample. This material is now in use for fission cross-section measurements.

The second sample cross-section was prepared by a 10-day irradiation of the same 50 grams of protactinium-231. This sample, about 1 gram of U-232, contains 0.7% U-233 because of the longer irradiation period. This material is now in use for total cross-section and capture cross-section measurements at the MTR.

Only 17 milligrams of uranium pure material is on hand in the research pool. However, we will obtain another 100 milligrams of the less pure material by reprocessing raffinates in the next few months, which will be available in the research materials pool and may be obtained by the usual procedures.

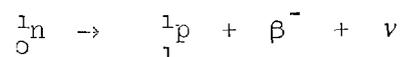
It should be added that we will be able to obtain about 10 milligrams of Th-228 per year by milking large samples of U-232 periodically.

ACCELERATOR-PRODUCED ISOTOPEs

J. J. Pinajian, ORNL

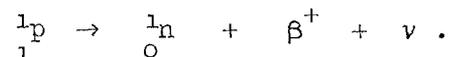
Webster's Third New International Dictionary defines coordination as, "to bring into common action" In this framework "Cyclotron Produced Isotopes" do indeed fit into this meeting because the cyclotron may be used to produce neutron deficient radioisotopes which in a small, but useful way, complement the nuclear reactor - making available more of a variety of radioisotopes.

In a naive sense, radioactive isotopes may result from the addition of a neutron or from the addition of a charged particle to a target nucleus (in our case, the charged particle is a proton). The former is generally accomplished using a reactor; in the latter case, a cyclotron or accelerator is used as a source of the charged particles. The radionuclides formed by increasing the neutron/proton ratio generally decay (or transform) back to a stable condition by having a neutron transform to a proton with the emission of a negative beta particle and a neutrino:



For those radionuclides resulting from an increase in the proton/neutron ratio, the transformation, again, tends to reverse the cause for instability,

and a proton in the nucleus will be transformed into a neutron, with the emission of a positive beta particle (positron) β^+ :



In some cases, nuclei which are unstable because of an excess of protons (neutron-deficient) will regain stability by capturing an orbital electron (ϵ):



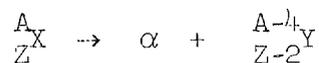
As a result of this process, characteristic x-rays will be emitted during the refilling of the orbital electron shells.

In the transformation processes described above, gamma radiation (γ) may or may not be emitted. These gamma rays, or photons, may undergo internal conversion whereby the transition between the two energy states of a nucleus is not evidenced by the emission of a photon. Instead the energy is imparted to an orbital electron, which is ejected from the atom.

For some nuclei, only gamma radiation is emitted for the de-excitation from a metastable or isomeric state. Such decay is termed isomeric transition (IT) and is characterized by no change in mass or atomic number.

Many radioactive nuclides decay by two or more modes, so that β^- , β^+ , γ , and ϵ transformations with associated γ emission and x radiation are not uncommon. The branching ratio defines the relative amount of each mode of decay.

For heavy nuclei, the transformation to a more stable configuration may take place by the emission of an alpha particle (α). Naturally radioactive nuclides of $Z > 82$ generally decay by α emission:



We no longer make irradiations in the ORNL 86-Inch Cyclotron which produce α emitting radioisotopes. The contamination problems associated with such endeavors are rather formidable and we tend to circumvent this sort of problem.

The principle reactions utilized for production purposes are:

<u>Reaction</u>	<u>Example</u>
p,n	${}^7\text{Li}(p,n){}^7\text{Be}$
p,2n	${}^{69}\text{Ga}(p,2n){}^{68}\text{Ge}$
p, α and p, α n	${}^{25}\text{Mg}(p,\alpha){}^{22}\text{Na}$ and ${}^{26}\text{Mg}(p,\alpha n){}^{22}\text{Na}$
p,pn - decay \rightarrow	${}^{58}\text{Ni}(p,pn){}^{57}\text{Ni}$ - decay \rightarrow ${}^{57}\text{Co}$

These reactions produce, of course, carrier-free activities so that the specific activity (curies/g) is dependent on the purity of the target material. By using isotopically enriched materials the yield of unwanted reaction products may be substantially reduced. We have been able to routinely produce ${}^{57}\text{Co}$ with specific activities of >6 curies/mg of cobalt (70% of theoretical) by exercising very careful control over our target materials and processing chemistry. High-specific activity ${}^{55}\text{Fe}$ has also been produced by perfecting manganese electroplating techniques specifically designed to eliminate iron impurities. Approximately two curies of ${}^{55}\text{Fe}$ has been produced with a specific

activity of 760 curies/g. Rubidium-84 for medical applications was produced from krypton gas depleted in ^{86}Kr , which decreased the production of the unwanted ^{86}Rb . The krypton, after irradiation, was collected for reuse in a Mionel thimble immersed in a liquid-nitrogen trap, and the radioactive rubidium was washed out of the chamber with hot distilled water.

Our cyclotron continues to be the prime source (indeed, for all practical purposes the only source) of neutron deficient radioisotopes. These isotopes give rise to 1) proton emission which result in 0.511 Mev annihilation quanta or 2) electron capture which results in characteristic x-ray emission.

Table I lists some of the more routinely produced isotopes from the ORNL 86-Inch Cyclotron, the targets used, and the experimental yields in mc/hr.

Table 1. Experimental Yields of Carrier-free Radioisotopes Routinely Produced
in the ORNL 86-Inch Cyclotron

Radioisotope	Flat-plate target		Capsule target		Window target	
	Material	Yield/hr, mc	Material	Yield/hr, mc	Material	Yield/hr, mc
^7Be	Li on Cu	30.0	LiH	16.0	-	-
^{22}Na	Mg	0.35	Mg plug	0.04	-	-
^{48}V	-	-	TiO_2	25.0	-	-
^{52}Mn	-	-	Cr powder	65.0	-	-
^{55}Fe	Mn on Al	8.0	-	-	-	-
^{57}Co	Ni on Cu	25.0	-	-	Ni foil	1.0
^{68}Ge	Ga on Cu	3.0	-	-	-	-
^{74}As	-	-	Ge powder	22.0	-	-
^{84}Rb	-	-	-	-	Kr gas	1.1
^{85}Sr	-	-	RbCl	5.0	-	-
^{88}Y	-	-	SrCO_3	5.0	-	-
$^{95\text{m}}\text{Tc}$	-	-	Mo tube	2.5	-	-
^{108}Cd	Ag	9.0	-	-	-	-
^{139}Ce	-	-	La_2O_3	1.2	-	-
^{181}W	Ta	13.0	-	-	Ta foil	0.5
^{195}Au	Pt on Cu	8.5	-	-	-	-
^{207}Bi	Pb on Cu	0.3	-	-	-	-

AMERICIUM CAPABILITY AT ROCKY FLATS

A. K. Williams, Rocky Flats

Americium-241 appears in Rocky Flats recovery streams as a decay product of ^{241}Pu .

Americium is recovered only from a precipitation stream where it follows in the filtrate. The filtrate is concentrated by evaporation and most of the Pu is separated by anion exchange. The ion exchange effluent, containing from 50 to 500 mg Am per liter and a like amount of Pu, is then concentrated and purified by ion exchange procedures which have been described by Ryan and Pringle in RFP-130. The basic process is the thiocyanate complexing process as reported in 1953 by Coleman, Penneman, and others. A simplified flow sheet is found in Fig. 1.

At the present time all shipments are made as americium oxide. Oxide is made by calcination of the oxalate at 800°C . The early shipments were chloride solutions, which presented some hazards in shipment and opening of the containers. Capacity of the present facility is approximately 360 g per year. Actual production since the first shipment in 1957 is found in Table 1. The reduction in Am shipped this past fiscal year reflects changes in production schedules and increased downtime of equipment. Purity of oxide shipped is found in Table 2. Note that many limits of detection are high. Cerium, lead, iron, and silicon are usually present in detectable amounts. These impurities are introduced into the process stream. The Am recovery line is shielded with 1/8 in. lead. Gloves impregnated with lead are used to minimize personnel exposure from soft gamma radiation. All process equipment is either PVC or glass.

A new Pu recovery facility is now under construction at Rocky Flats. Included in this is a larger Am processing line which will have a capacity of 3600 g per year on a 5-day, 3-shift schedule, and 5000 g per year when expanded to 7 days. This new facility will use the same process. It will not have provisions for recovering Am from other streams which are now discarded to waste. These streams, which are highly salted and contain 1-5 mg Am per liter, discarded approximately 600 g in FY 62 and 880 g in FY 63. Discard rates for FY 64 so far have averaged 72 g per month.

Processing costs in our present facilities have averaged \$1000-1500 per g. When the new facility is complete, estimated processing costs are

Figure 1
Rocky Flats Americium Recovery Process

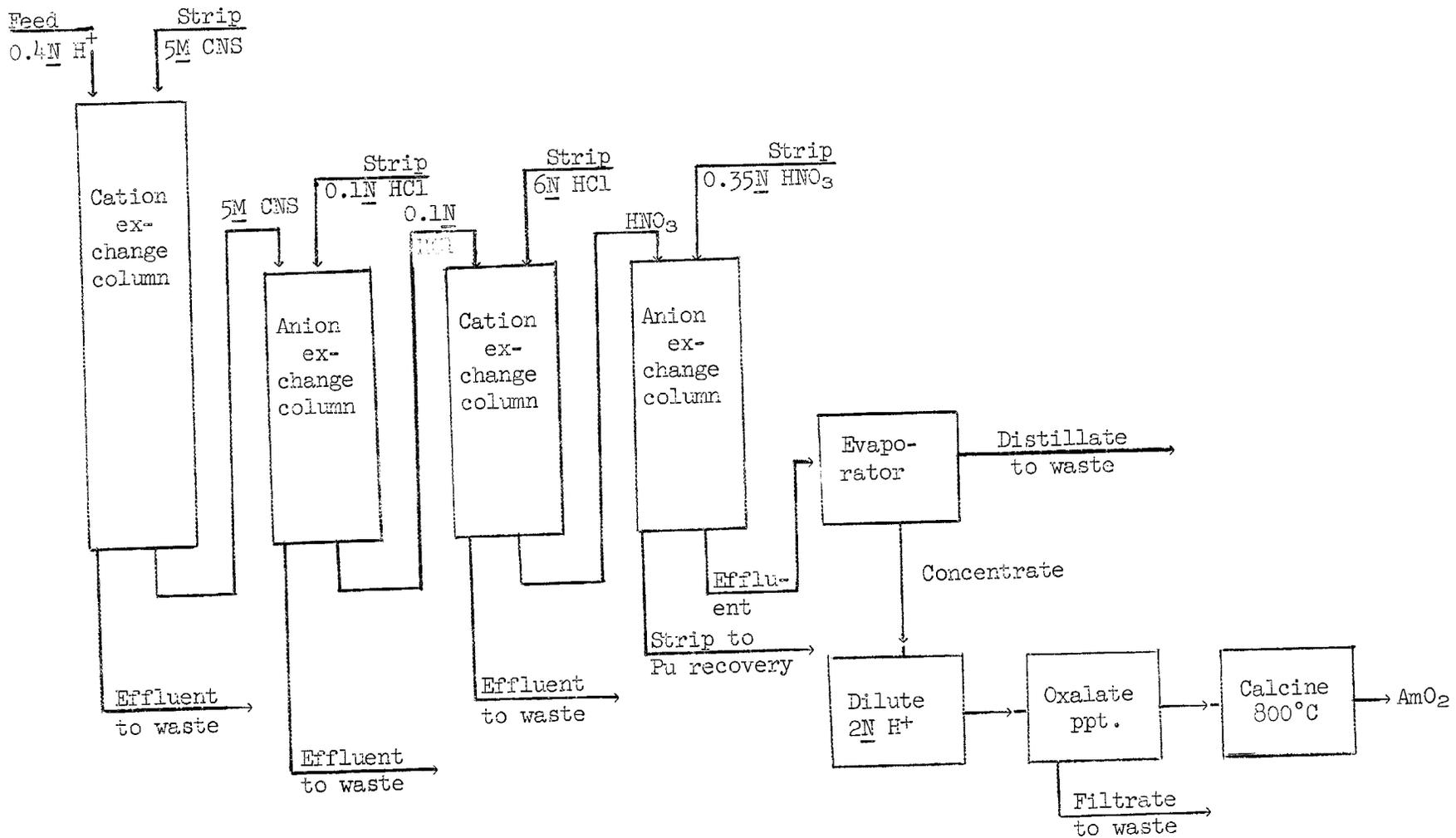


Table 1. Americium Production, Rocky Flats Plant

Fiscal year	Americium, g
1956	1.9
1957	9.7
1958	17.6
1959	171.6
1960	213.3
1961	239.1
1962	359.7
1963	124.0

Table 2. Americium Oxide Spectrographic Impurities, ppm

Detectable impurities	< 500	< 100	< 50	< 10	< 5	< 1
Al-800	Cs	As	Au	Bi	Lu	Be
B-10	Nb	Nb	Dy	Co	Se	
Ba-50	Sm	Cd	Eu	Mn	Y	
Ca-600		Ge	Ga	Mo	Yb	
Ce-1000		Hg	Gd	Sn	Zr	
Cr-500		Li	Hf	Ti		
Cu-50		Na	Ho	V		
Fe-1000		P	In			
K-100		Pb	Ir			
Mg-1000		Rb	La			
Ni-100		Sb	Pd			
Pu-1000		Sr	Pt			
Si-500		Ta	Re			
		Te	Rh			
		Rh	Ru			
		Tl	Tb			
		W	Tm			
		Zn	V			

\$140 per g when operated at the design capacity of 3600 g per year. This is assuming that Pu recovery costs would not be charged to the Am product.

In order to recover as much Am as possible, all Pu rich in Am could be sent to recovery for chemical separation. The costs for Pu processing would then be charged to Am. These costs are estimated at \$500 per g Am for the new facility. The cost of Am can fluctuate from \$140 per g to over \$700 per g, depending on Pu processing charges.

Rocky Flats has a sizable quantity of residues on hand which are estimated to contain approximately 1200 g Am. It is estimated that 950 g could be recovered in the three years between FY 65 and FY 67, provided we are successful in developing a new recovery process and that new equipment would be available.

Process development is proceeding in two areas at Rocky Flats. The first is recovery of Am from these dilute, highly salted waste streams. The process currently under study is solvent extraction of both trivalent and hexavalent Am. The second is a molten salt extraction process, where Am dissolves more readily in molten chlorides than Pu. This was found in our Pu electrorefining work, and we feel that a process of this type might be more economical than wet chemical processing. We also hope to reduce costs to the tens of dollars per g range rather than the current cost of thousands of dollars per g.

Our present development facilities consist of a 200-ft² laboratory, containing 15 ft of glove boxes shielded with lead. We are planning on installing an additional 15 ft this fiscal year. This laboratory is presently staffed with one chemist, full time.

Our analytical facilities are the same as those used for Pu. At the present time, we are equipped to run spectrographic impurities and radiochemical assay on solutions. We are currently attempting to improve our analytical methods for both impurity analyses and assay. Among impurity analyses being developed are the polarographic determination of lead, fluorimetric determination of uranium, and spectrographic determination of thorium after an ion exchange separation.

Assay procedures being developed include low-geometry alpha counting, titrimetric methods using EDTA, and colorimetric methods measuring the trivalent Am peak at 502 mμ.

THE PROPOSED ORNL RADIONUCLIDES SEPARATIONS LABORATORY

A. F. Rupp, ORNL

While my scheduled subject is on a particular facility that we are proposing to build (Radionuclides Separations Laboratory - "RSL"), I should like to talk for a few moments in more general terms about the need for the kinds of special research materials that one would produce in such a facility. In brief, we consider that a worthwhile goal is the ultimate production of usable quantities of all possible nuclides as nearly chemically and isotopically pure as possible, whether the nuclides be stable or radioactive. We have ample evidence here at the ORNL Isotopes Development Center that experimenters have great need for these special research materials.

In nuclear physics, thin films only a few atoms thick are used for particle interaction and decay studies; in almost any case where nuclear physics measurements are made, the experimenter is interested in defining his materials as absolutely as possible. In chemistry, ordinary tracer dilutions usually suffice and high specific activities are not required; however, modern chemistry has come to rely greatly upon physical methods--measurements that are made on chemicals at high dilution, in thin films, on surfaces, or in crystal lattices require high radioisotope purity. Biomedical research has great need for high specific activity materials, since biologists prefer to disturb cellular chemistry as little as possible and therefore object to introducing any more foreign chemicals than necessary. In complex biochemical systems, the radioisotope tagging of certain positions of large molecules is difficult if receptor positions are taken up by the inactive atoms. Here again, a radioisotope preparation that contains a high proportion of the radioisotope atoms desired is helpful.

In using the radioisotope preparations for sources of radiation and heat, one again encounters specifications demanding high isotopic purity. Point sources of gamma and x rays are preferred for radiography; alpha and beta radiation sources must be made in thin layers to prevent self-absorption of the radiations; heat sources to generate electric power in the most advanced thermionic conversion systems must have the highest possible power density. With the rising level of sophistication of research and development work utilizing radioisotopes, one may expect increasing demands for very high specific activity materials.

For the separation of stable isotopes, we use the electromagnetic separators (calutrons) and thermal diffusion columns (principally for the rare gases, such as Kr, Ne, Ar). Other possible methods are known, but for relatively small-scale production, these two methods are clearly the most versatile and practical.

Several other approaches are available to us in producing pure radioactive nuclides, or even some stable nuclides whose precursors pass through relatively short transmutation decay processes. The most important are as follows:

1. Direct transmutation reactions - n,p ; n,α .
2. Use of decay chains - n,γ ; (β^- , β^+ , α , EC^-); n,f ; (β^-,α).
3. Burnout of target atoms.
4. Isotopic separation of radioactive nuclides.

Direct neutron transmutation reactions are few in number and well known--e.g., ^{32}P from sulfur or chlorine; ^{14}C from nitrogen; ^3H from lithium. Transmutations during decay after n,γ reactions are also familiar--e.g., $\text{Te } (n,\gamma,\beta^-)^{131}\text{I}$; $\text{Xe } (n,\gamma, EC)^{125}\text{I}$. Transmutations during and after the fission process are very numerous.

However, as I pointed out in a paper given at the Japan Radioisotopes Conference in May (1963), there remain many radionuclides that cannot be made pure by transmutation processes. Therefore, we are examining two routes to these more inaccessible objectives, namely, target burnout in ultra-high-flux reactors such as the HFIR and physical isotopic separation of radionuclides. Unfortunately, not very many pure nuclides can be made by target burnout, for one reason or another (too long or too short a half-life, too low a neutron capture cross section, etc.). We are left with many nuclides where the last resort is physical separation of the isotopes. Since the electromagnetic process is by all odds the most versatile method of isotopic separation, we are designing a \$5.5 million facility around several such machines, adapted for remote control work with alpha-beta-gamma-neutron emitting nuclides.

There is need for the pure radionuclides as special research materials, but the manpower and money required to obtain them is very considerable. We are preparing a comprehensive report covering all aspects of this program background, need for the materials, conceptual designs of the calutron equipment, auxiliaries, and plant. The report will be issued in the spring of 1964.

STABLE ISOTOPE PROCUREMENT

P. V. Arow, ORNL

Sales

There are approximately 250 enriched stable isotopes available on a sale basis, and they have a wide range of prices calculated to realize full cost recovery. There is no profit increment represented in the prices. The enrichment of these materials is accomplished by electromagnetic separation and by other methods, conveniently designated as non-electromagnetic separation. We usually refer to them as EM and non-EM isotopes.

The extremes of the price range are \$0.05 per mg of element for 25-50% ^{207}Pb to \$1400 per mg of element for a 6-mg sample of ^{40}K enriched to 84.1%. This sample is not actually a sale item, even though it does have a valuation.

In our "Catalog of Radio and Stable Isotopes," the second section (composed of pink pages) is devoted to the stable isotopes which are generally available. Listings are by element and isotope, with natural abundances, enrichment ranges used for pricing, the prices (in effect at the time of revision, April 1963), and a class coding to indicate the usual quantities available. Prices of the stable isotopes change frequently because of constant new production. As a result, many of the prices in the catalog are obsolete. In addition, all enrichment ranges are not necessarily available.

When an order is received for materials on which there has been a price change, the purchaser is voluntarily given the benefit of lowered prices. If certain prices have increased, the purchaser is advised of the new price so that he may determine acceptability or make an alternate selection before shipping. Prices of stable isotopes generally apply to milligrams or grams of element, not to the amount of isotope. For example, ^{57}Fe enriched > 85% is priced at \$6 per mg of element. On this basis, an order for 10 mg of ^{57}Fe enriched 91% would normally be shipped as 10 mg of Fe for \$60 and would contain ~ 9 mg of ^{57}Fe .

The following are problems which we frequently encounter in orders for stable isotopes:

1. Orders are submitted on the purchaser's own order form only, instead of on the required Government form. (The company's own order form and our required Government form sometimes differ in the information given.)
2. If the isotope is inventoried as a compound, it should be clearly indicated whether the amount desired is element or compound (e.g., 50 mg of Ba, as $\text{Ba}(\text{NO}_3)_2$, > 85% ^{136}Ba).
3. Try to be sure that the isotope listed has the correct enrichment and price for that isotope.

The method of purchasing stable isotopes by a Federal agency or by an AEC cost-type contractor is to send an order on Form AEC-375. All organizations other than Federal agencies send orders on Form AEC-391. These forms are used for purchasing either radio or stable isotopes. Note, however, that licensing by the AEC is not necessary when purchasing stable isotopes, and the related section of the order form may be disregarded.

Stable isotopes are shipped once each week, with Thursday being the target day. Orders should be received by noon of Tuesday for inclusion in that week's shipment. We have been averaging 240 shipments of stable isotopes per month, including both sales and loans.

With regard to our catalog, a number of copies are available here, but if you would rather not carry a copy, we will be glad to mail one to you if you will give us your name and address. The catalogs are available at no cost to the user.

Loans

In the first 10 years of the stable isotope separation and distribution program (1946-1956), electromagnetically separated stable isotopes were made available solely on a loan basis. They were distributed after authorization by the U. S. Atomic Energy Commission. In 1956 a transition was made to provide stable isotopes chiefly on a sale basis, and at that time authorization by the Commission became no longer necessary.

In conjunction with the sales provision, a coordinate measure provided that, by satisfying certain regulations, the loan of certain electromagnetically separated stable isotopes to domestic users would still be permitted. These regulations provide that:

1. Loans will be made for research purposes only. The loan must be deemed justifiable on the basis of the proposed research use. This use must be of such nature that there is little likelihood that the material will be lost, destroyed, diluted, or contaminated.
2. Loans can be authorized only for rare and expensive items and may be made only to institutions, not to individuals.

In order to consider a request for loan of isotopes we must obtain from the requester a reasonably detailed statement of the proposed usage, the amount of material needed, and the approximate length of time for which it will be required. From experience, it is generally pointed out to the prospect that, if activation of materials is to occur, any resulting nuclides should not have half-lives greater than about three months and that the amount of activity should be on the order of a few micro-curies.

The following provisions must be adhered to by the borrowing institution:

1. It must agree to be financially responsible for any loss of value by dilution, contamination, or loss of the material incurred while the loaned item is in its possession. The user must agree to return the isotope in inventory form and purity or reimburse ORNL for costs of our reprocessing.
2. The prospective borrower negotiates a suitable loan period for the proposed research. Loans may be granted for periods of 3, 6, 9, or 12 months. Extension of the loan period will be considered upon the borrower's request prior to the loan's expiration date.
3. The borrower agrees to either publish results of research performed with these materials or make the results available to the U. S. Atomic Energy Commission upon request.
4. At the end of the loan period, the borrower returns with each isotopic sample, two copies of Form OR-629, Certificate of Isotope Accountability. After the sample has been analyzed, the user will be billed for any loss, dilution, and/or chemical reprocessing necessary.

The following charges are authorized for stable isotope loans:

1. An analysis and handling charge of \$100 for each sample loaned is billed at the time of shipment.
2. A use charge of $4\frac{3}{4}\%$ per annum of the sale value of the item loaned is billed to non-Federal agencies for materials

loaned from active inventory. The minimum use fee charged is for a period of three months. All Federal agencies are excused from payment of the use fee.

The cost of special customer services performed by ORNL on loaned materials (such as conversions or the fabrication of particular shapes) is, of course, charged to the user, just as in the case of the sale of isotopes.

3. If a loaned item has not been returned at the expiration date of the loan period and a request for extension has not been received, the borrower is subject to billing for the full sale value of the material, and the transaction may be treated as an ordinary sale.

There are 26 isotopes which are specified as being available for sale only. Most of these materials are of relatively high natural abundance and would represent relatively low concentrations as separated samples. A few examples of the items available for sale only are: ^{137}Ba , n.a. ~ 11%, any enrichment < 27%; ^{52}Cr , n.a. ~ 83%, any enrichment < 97%; ^6Li , n.a. ~ 7%, all enrichments; ^{120}Sn , n.a. ~ 32%, all enrichments. Any variation from a sale basis for any of the 26 isotopes in question would be a matter for individual consideration by the AEC.

Requests for loans of stable isotopes are made by means of the same purchase order form that is used for buying isotopes, i.e., Form AEC-375 for Federal agencies, and Form AEC-391 for all organizations other than Federal agencies.

In general, loans are not made to overseas users. Any deviation from this regulation would be at the discretion of the AEC.

RESEARCH POOLS

F. N. Case, ORNL

Several pools of isotopes specifically assigned for research uses are maintained by ORNL for the Atomic Energy Commission, Division of Research. These materials are available to research programs when the use is approved by the Division of Research.

Cross Section Pool

The Cross Section Pool (CSP) is an inventory of stable isotopes, preparation of which is scheduled jointly by the Isotope Development Center, the Division of Research, and the Cross Section Committee. Stocking and maintaining the CSP inventory is the concern of the Calutron Separation Program at the Isotope Development Center. Samples may be obtained from the CSP by communicating directly with George Rogosa, USAEC, Division of Research, who will review the experimenter's intended use so that it can be determined, while considering the data to be obtained, how the experimenter's requests can best be met.

In preparing various CSP samples, other isotopes of the element are often collected. Thus, if pool requirements for these isotopes have been met, the excess is placed in an active stable isotope sales inventory. These items are then offered to any purchaser through the normal isotope sales program.

Some research programs requiring samples of enriched isotopes may obtain them on a loan basis from the sales inventory. Loans are limited to samples that are rare and expensive, and the borrower must agree to be responsible (at catalog prices) for any losses. The CSP loan activity is best illustrated by Fig. 1. The solid line shows the number of loans made from the pool each year. Each separated sample loaned is counted as one loan. The broken line shows the accumulated number of outstanding loans. The total inventory value of outstanding loans is \$5,702,289.90. Fig. 2 shows the growth of loans from the sales inventory. The outstanding loans have a value of \$1,997,879.25.

The Isotope Development Center provides a number of services in association with the CSP and Research Pool materials. Often the experimenter

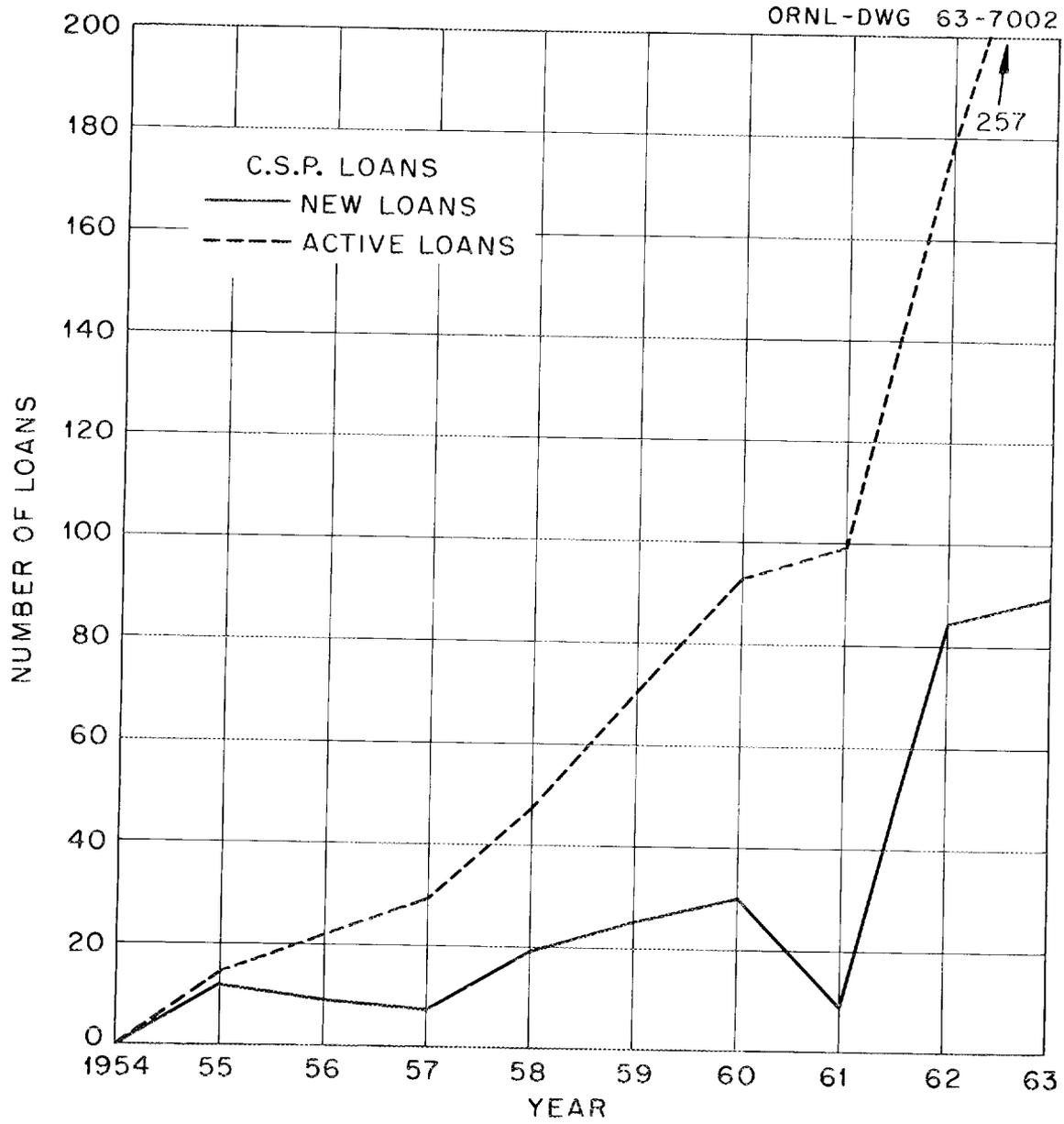


Fig. 1

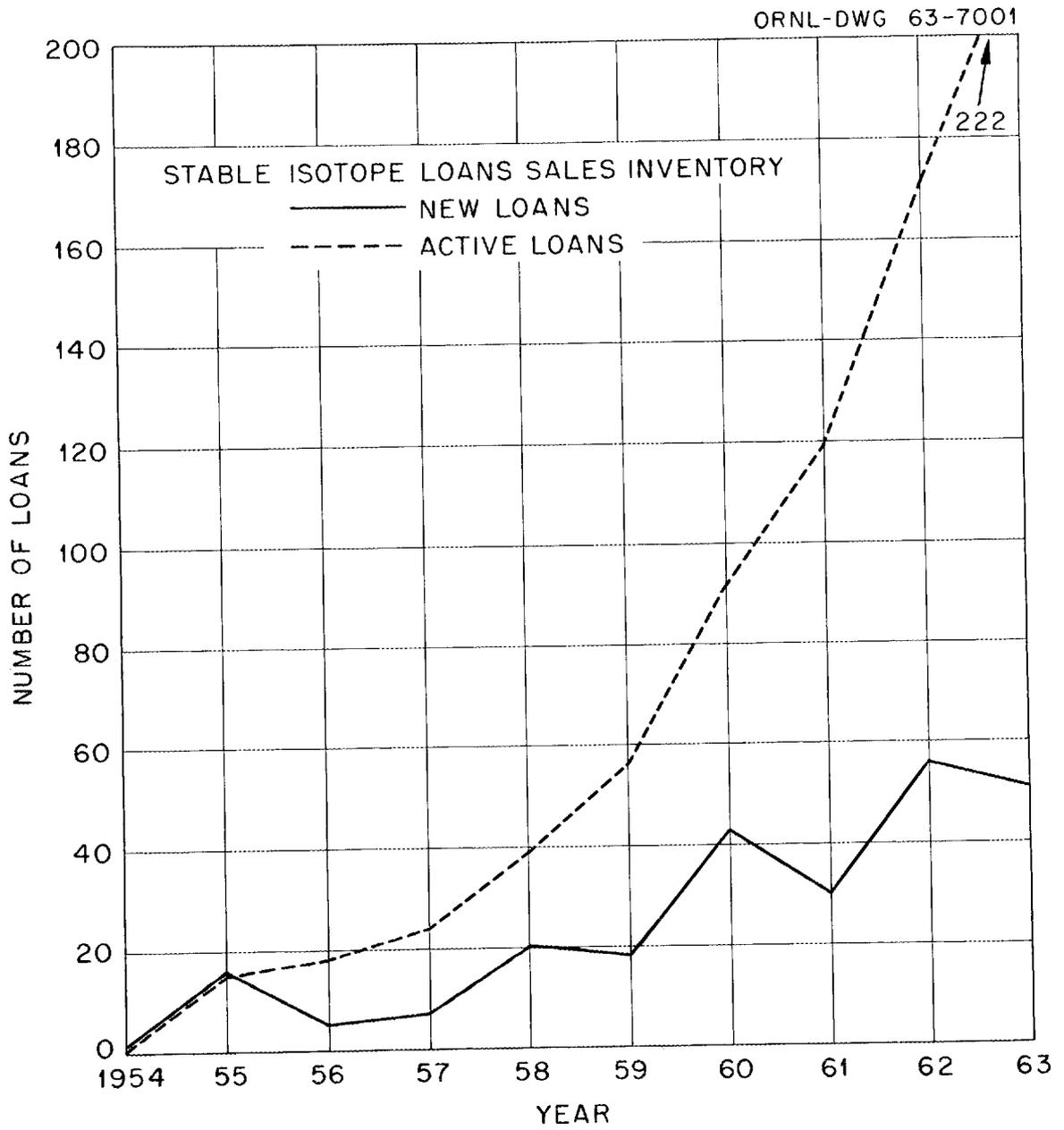


Fig. 2

requires that the isotope be in a form other than the standard inventory compound. Conversion to a special chemical form and fabrication into various geometries such as foils, cylinders, or plates can be provided to the experimenter. Such services are reviewed for feasibility, conservation of the isotope, and effect on the scheduled use of the isotope when more than one experiment is involved. Thus if two experiments can be performed using a certain fabricated shape, those experiments would be scheduled to best utilize the fabrication effort.

Cross Section Pool inventory information is distributed by the Division of Research. Each month, the Isotopes Development Center supplies records to the AEC concerning transfers of pool materials. These transfers include loans, loan returns, and new inventory samples.

Heavy Element Research Pool

The Heavy Element Research Pool is maintained for the Division of Research and continues its identity as a separate pool because of special handling problems. The pool is primarily concentrated at ORNL; however, other AEC facilities may be involved in preparing samples. As a consequence, the physical inventory might be located at their facilities.

Calutron separation of heavy elements began nearly 16 years ago, with the preparation of samples of highly enriched ^{238}U , known at that time as sulfur Q. Highly enriched ^{234}U and ^{235}U samples were prepared following the sulfur Q program; next, high-purity ^{236}U and ^{233}U were separated. About 10 years ago the Pu isotopes were separated in the calutron in order to provide enriched samples of ^{240}Pu and ^{241}Pu for obtaining physical data which would permit decisions relative to Pu production management. (The current Pu program was discussed earlier.)

From time to time, a number of specific isotopes have been placed into the Research Pool, thus reserving them for Commission-approved experiments. In 1948 two batches of ^{14}C (~ 1000 mc of 48% ^{14}C and 1600 mc of 80% ^{14}C) were placed in this group. Technetium-99 was effectively placed into research pool status by putting a maximum sales quantity limit on it. Non-AEC facilities wishing to procure greater than 10 g per year must obtain approval from the Division of Research. AEC facilities are limited to 100 g per year without specific approval. The supply situation is much improved,

and approval for larger quantities is usually provided without difficulty. Approximately 26 kg are in process. Other isotopes controlled by limiting sales to a specific quantity or per cent of inventory are ^{10}B , ^{11}B , ^6Li , ^{241}Am , and ^{237}Np . These quantities may change from time to time, depending upon the balance on hand and anticipated requirements.

Allocation from the pool is made by both the Division of Research and Development, ORO, and the Division of Research, Washington. Experimenters wishing to obtain samples from the pool should contact the ORNL Isotope Sales Department so that it can be determined whether or not samples meeting experimenters' requirements are in the pool. If samples are in inventory, the Sales Department will refer the experimenters either to T. R. Jones, AEC, Washington, or to Herman Roth, AEC, ORO, depending upon the sample involved.

The inventory of isotopes distributed from the Heavy Element Research Pool contains $^{230}\text{Th}^*$, ^{231}Pa , ^{232}U , $^{233}\text{U}^*$, $^{234}\text{U}^*$, $^{235}\text{U}^*$, $^{236}\text{U}^*$, $^{238}\text{U}^*$, ^{237}Np , ^{238}Pu , $^{239}\text{Pu}^*$, $^{240}\text{Pu}^*$, $^{241}\text{Pu}^*$, $^{242}\text{Pu}^*$, and ^{241}Am . Examples of some of the enrichments available are as follows:

Isotope	Batch no.	Isotopic purity, %	Weight, g	Chemical form
^{241}Pu	Pu-241-102A	93.27	0.4476	PuO_2
^{242}Pu	Pu-242-103A	91.47	0.2069	PuO_2
^{238}Pu	Pu-238 M2	80.96	4.42	PuO_2
^{239}Pu	St-101	99.979	0.175	PuO_2
^{240}Pu	Pu-240-101A	98.00	10.906	PuO_2
^{235}U	F2S-22(c)	99.994	0.0269	U_3O_8
	F2S-22(f)	99.888	14.81	U_3O_8
^{236}U	U-236-E-R	99.44	0.005	U_3O_8
	IMR-236-3B	95.00	0.033	U_3O_8
^{238}U	Q-505	99.9999	0.525	U_3O_8
	K-25-36(b)	99.9996	29.98	U_3O_8
^{233}U	V3J-43235	99.9963	0.0563	U_3O_8
	ETA-R2	99.76	50.70	Metal foil
^{234}U	HR-234-1B	98.27	0.0117	U_3O_8
^{232}U	ORNL 1-4	99.1	0.0169	NO_3 solution

* Calutron separated.

The Laboratory's facilities are quite diversified, as described by earlier speakers. Both radio and stable isotopes can be processed by the Isotopes Development Center, thus allowing a great deal of latitude for the experimenter.

RESEARCH SAMPLES PROGRAM

L. K. Hurst, ANL

Argonne National Laboratory is represented at this meeting primarily in the procurement role and as a user of the special research materials and services which perhaps should be made available by USAEC production organizations rather than research laboratories. It is not Argonne's intention to produce materials; although some new items are prepared as research efforts result in discovering new compounds, isotopes, elements, and production techniques or, as these efforts require unique materials in the course of the studies. Many of you are aware of past achievements by the Argonne staff in the discoveries of tritium labeling of organic compounds, the biological synthesis of ^{14}C compounds and deuterated compounds, the fluorides of the "inert" gases, methods to produce ultrahigh-purity uranium and plutonium metals, methods to prepare the phosphides and sulfides of uranium, the development of nuclear reactor systems, and so on. In practically all cases, the products of these studies are prepared for others to perform complementary research until such time when the techniques for production are understood sufficiently enough to permit preparation elsewhere to be accomplished successfully.

The Argonne Laboratory is large and has many interests. It is one of the nation's principal facilities for research in nuclear energy and for the development of the atom as a servant to mankind. Briefly, it has 14 divisions of research and development personnel representing all disciplines in the physical sciences. Approximately 1,000 scientific staff are directly employed in its programs. While a prime function is the design and development of nuclear reactors, nearly half the Laboratory's effort is devoted to basic research performed for the sake of research.

Argonne's physicists, chemists, biologists, and metallurgists, performing basic research, need instruments, tools, and materials with which to conduct their work. They already have a large array of sophisticated research instruments and analytical tools such as Van de Graaff accelerators, a linear accelerator, cyclotron, the zero gradient synchrotron, the research reactors - CP-5, Juggernaut and Janus, 100-in. spectrometer, neutron and beta spectrometers, and others too numerous to mention. By

respectful appreciation of the needs, the Atomic Energy Commission is increasing the stock of fine equipment. The Argonne Advance Research Reactor, the Faret, the ZPPR, and the 12-foot bubble chamber are only a few of the items now on the drawing boards.

You might ask what all this has to do with this meeting. Well, it is an attempt to impress upon you that the array of equipment at Argonne National Laboratory is so complicated and so expensive that much of it is not duplicated in this country. It is costly in terms of construction costs, operating costs, and the scientific manpower which has been accumulated to use it. Both the laboratory staff and the personnel of the Midwestern Universities with whom there is close cooperation are selected for and oriented toward using this fine inventory of equipment in research rather than being involved in producing special samples needed by them and their colleagues.

We are here today to learn about the capabilities of the various production groups and to identify our needs, for it is becoming increasingly difficult to find the special articles needed by this staff. Considerable success toward these objectives has already been achieved by the expansive discussion held so far. We do implore you though, regardless of the satisfaction felt, to communicate, on a regular basis, the current information about materials and services available and their technical specifications. In addition, we ask that the producers continually seek methods and techniques to purify more extensively, in element form, these rare and exotic materials and to find as broad a base as possible over which to distribute their costs. We know special work is expensive, but at the same time one can make or break a very costly experiment by failing to completely purify to the extent required or by assessing the entire cost of production and preparation to an individual program.

Obviously, Argonne's interests will not remain constant; however, roughly speaking, there is interest in obtaining high-purity metals, both natural and separated isotopically. These interests vary from gram to kilogram quantities. The alkali earths, the rare earths, particularly the odd-odds, and high-purity metals of other stable isotopes are specially desired. Reactor control materials, lithium and boron - again well purified and of course, isotopically separated - are needed in large quantities.

Neutron threshold detectors such as pure ^{236}U , ^{236}Np , and ^{242}Pu are almost impossible to find. And finally, somewhat out of the metals class, we need reasonably large volumes of essentially tritium-free heavy hydrogen.

Earlier, the point was made that Argonne is well equipped for research which its staff and those from the Midwestern Universities are conducting. Clarification is necessary to assure you that this equipment is available for any AEC-sponsored work within the limits of its capabilities and capacity and that the staff of any of the other laboratories or facilities of the Commission who have a bona fide need for these specialized services should make known their interests. These facilities are offered to complement, rather than compete with, others; in fact, encouragement is given to obtain services from other sources if it is at all possible, because considerable competition for the current output of the equipment exists between resident staff members.

Some of these specialized instruments in which others may have interest include the 60-inch cyclotron, the 100-inch mass spectrometer, the electromagnetic isotope separator, the synchrotron, the Janus reactor, and the chemistry high-level hot laboratory. While specific details about availability, capability, and operating policy can be obtained directly from the operating staff, the facilities are briefly described for purposes of acquainting you with their general characteristics.

The Argonne cyclotron is a constant-frequency, 60-in. machine which has been in operation since 1952 for extremely diversified research interests in heavy-element chemistry, nuclear activation studies, nuclear scattering, solid states physics, and others. Deflected beams of deuterons, helium ions, protons, and neutrons of broad energy spectrum are constantly in demand. The typical deuteron operating data are: a resonant frequency of 11.2 Mc/sec, 240 kv peak dee-to-dee voltage, 75 kv dc deflector voltage, 600 μa internal beam, and 200 μa deflected beam. The tuning of the cyclotron is accomplished by varying the radio frequency with the magnetic field held constant.

The 100-inch-radius mass spectrometer is a double-filament surface ionization instrument using a combined electrostatic analyzer and magnetic analyzer in a Mattauch-Herzog arrangement to produce focusing of the ion beam in both direction and velocity. The transmission of ions is $> 95\%$.

over-all for both the ion source system and the analyzer. The resolution is 10^4 . The spectrometer is normally used for high-sensitivity isotopic analyses of very small amounts of material such as products of nuclear reactions produced by accelerator bombardment and for precision mass measurements.

The electromagnetic isotope separator is basically a large 90° magnetic deflection mass spectrometer capable of maintaining good resolution and good separation between neighboring isotopes while focusing moderately large beam currents. It can cover the entire mass range and can handle materials in elemental or compound form as solids, liquids, or gases. The ion beam with an energy variable in the range 3-70 kv and intensity up to 1 ma can be focused to a fine line (0.5 mm \times 20 mm or more long), or to a small spot of 2-3 mm dia, or defocused to a much larger area. Enrichment factors for neighboring isotopes of 10^3 - 10^4 are obtainable, and the efficiency of separation is generally in the range of 1-10%.

The zero gradient synchrotron is the first multi-Bev particle accelerator in the Middle West. It is one of the 3 most powerful proton accelerators in the world and currently has the most intense beam (up to 5×10^{13} protons per pulse). It exceeded its design energy of 12.5 Bev upon startup. High-energy physicists from universities and Argonne are employing this instrument for basic nuclear research on subatomic particles with scheduling arranged following recommendations from the ZGS users group.

The Janus reactor is a heterogeneous light-water-moderated reactor of the tank type, having radiation faces delivering different neutron intensities to rooms for low-level and high-level irradiations. Doses on the high-level side will range from 1 rad per min to 100 rads per min and on the low-level side, will range from 0.1-50 rads per week. Other special features, such as movable converter plates over each face to produce fast neutrons, neutron attenuators, and gamma shields, exist.

The chemistry hot laboratory is a cave-complex constructed of monolithic concrete and consisting of 2 floors of shielded cells. The walls on the lower floor are 4 ft thick with magnetite ore filling on 3 of the 4 cells to provide shielding against 10^6 curies of 1-Mev gamma activity or a point source emitting 10^{12} fission neutrons per sec. Walls on the second floor are 28 in. thick -- again with magnetite concrete in 3 of the

8 cells to provide shielding against 1,000 curies of gamma activity or a point source of 10^9 neutrons per sec. The other cells can accommodate ~ 30 curies of gamma activity. The facility is employed primarily for radiation chemistry and heavy-element research.

RESEARCH MATERIALS INFORMATION CENTER

J. W. Cleland, ORNL

The Pure Materials Program of the Solid State Division was organized to develop the necessary techniques as required for the growth of ultra-high-purity and controlled-impurity research specimens of immediate and long range interest to ORNL, other AEC installations, and other AEC contractors. The present program directly involves eleven people at ORNL; however, it also requires the direct assistance of many other research and service groups.

The actual choice of materials has been made, for the most part, on the basis of an expressed desire on the part of individual researchers and on the basis of the potential gain toward a fundamental understanding of solids. The ultimate range of certain properties in many materials can be established only by means of experiments that are conducted on pure specimens, usually single crystal, or on samples that contain a known type and amount of chemical impurity.

Detailed information concerning the availability of research materials has always been difficult to obtain. Open literature publications seldom indicate the source and purity of starting materials, further purification, methods of crystal growth, final purity, and method of assay and analysis of the end product. The Research Materials Information Center was therefore established as a part of the Pure Materials Program to collect and provide information on the purification, production, characterization, and availability of research quality materials to both producers and users. A companion listing is also made of those materials that are not currently available but are desired by various investigators. T. F. Connolly is in charge of the Research Materials Information Center.

Three distinct phases are usually required for any research material of interest. These are (1) initial purification, (2) crystal growth, and (3) evaluation of the end product. The present program includes such materials as KCl, HgS, Cu₂O, isotopic LiF, UO₂, MgO, CaF₂, BeO, ThO₂, solution grown quartz, beryl, TiO₂, and others. Most of the actual work is reported in the annual progress reports of the Solid State, Reactor Chemistry, and Metals and Ceramics Divisions and in the monthly status

and progress reports of ORNL. Special analytical techniques, such as mass spectral analysis, neutron activation analysis, and microspectroanalysis have been developed in the Analytical Chemistry Division and are important for the initial purification and analysis of the end product. One must also rely heavily on physical measurements of electrical, optical, magnetic, thermal, and structural properties in any evaluation of the end product. It must be emphasized that close cooperation is required between those who analyze, grow, characterize, use, and receive or provide information on these materials, and this cooperation must transcend any group, division, or individual laboratory. It is also evident that purities, dislocation content, crystal size, crystal perfection, special properties, etc. are constantly being altered or improved and that one must depend on some such mechanism as an information center for the current status for any particular material.

The true role of the Research Materials Information Center is to extend the present cooperation between crystal growers and crystal users, as it already exists at ORNL, to a national or international basis. Equipment has been purchased and installed that will conduct an automatic search for coded entries that have been placed on film. More than 1,000 data sheets on a wide variety of materials have already been received from about 500 individuals representing more than 60 research groups. These data sheets have been coded as to material, form, dimension, orientation, impurity content, method of growth, analysis, intended use, etc. Any future change or improvement in the material is added to the data sheet and is also coded into the film; hence, the requester of a desired material that is not presently available will be notified if such a material becomes available at a later date.

Background information on materials, including purification, analysis, production, and evaluation, is also being collected and coded. A thesaurus of more than 300 coding terms has been established under such general topics as (1) Type and Form of Material, (2) Crystal Structure, (3) Growth Technique, etc. This thesaurus contains such specific coding terms as Bridgman, condensation, Czochralski, deposition, deBoer, flame fusion, hydrothermal, Kyropolous, Van Arkel, and about 60 others under (3) Growth Techniques, alone. The open literature from 1957 to the present has

already provided about 9,000 pages of abstracts, title pages, and documents as searched for (3) Growth Techniques, alone. This material has been coded on four 100-ft rolls of 16 mm film, where each film can be searched for a particular set of coding terms in about 15 sec.

One of the advantages of this technique is that the searcher can select his own coding terms from the thesaurus (e.g., ALSb, single crystal, 5 N purity, 100 orientation, thin film, or float zone growth). He can then scan each data sheet, abstract, title page, or entire document as the machine searches, and he can obtain a hard copy of any of the above directly from the machine if it is of particular interest. I hope that our visitors will find time to visit the Information Center, Room G-56, Building 4500-S, and that they will also pick up an ample supply of blank data sheets.

The present status of the Research Materials Information Center has been summarized in the first of what is intended to be a series of aperiodic bulletins. The first bulletin also contains three articles that describe the purification, crystal growth, and comparative analyses for KCl and isotopic LiF and an initial listing of available and desired materials. It is recognized that this listing is woefully inadequate; however, this was a first attempt. We are currently working on a second bulletin, which will also contain several articles on the growth of metal crystals. Any suggestions for yet future articles that might be of interest would be greatly appreciated.

I would now like to discuss some specific examples of work in the Pure Metals Program of the Solid State Division. T. H. GeBalle and associates of the Bell Telephone Laboratories obtained 17 g of 95.8% enriched ^{74}Ge from the Stable Isotopes Division of ORNL about 6 years ago. This material was zone refined and used to grow a very high-purity n-type Ge single crystal ingot. GeBalle employed this crystal to demonstrate the profound effect of isotopic scattering on low-temperature thermal conductivity. D. K. Wilson of the Bell Telephone Laboratories used a portion of the same crystal at a later date to ascertain the spin-lattice relaxation time in Ge that contained a known amount of a single chemical impurity, arsenic, as introduced by thermal neutron irradiation and subsequent transmutation in the LITR (Low-Intensity Test Reactor). His experiments

suggested that the original mass spectrometer analysis of the original ^{74}Ge stable isotope was in error. A second analysis, utilizing barium fluoro-germanate (BaGeF_6), showed that the ^{74}Ge content was 96.75% instead of 95.8%, which was in much better agreement with the spin resonance line width results. We have used portions of this same crystal to study (n,γ) recoil damage, where the actual recoil atom can be identified as a donor-type As chemical impurity after transmutation. This is one example of a great deal of experimentation on 17 g of a pure isotopic material.

C. T. Butler and J. R. Russell of the Solid State Division have grown several single crystal boules or ingots of KCl that are superior in quality and more uniform than any commercial material. This work has required several analytical techniques, including further purification of initially "spec-pure" starting materials, and many consecutive analyses of completed crystals.

Transparent single crystals of Cu_2O of moderate quality are being produced by the Virginia Institute of Scientific Research under contract, and M. J. Smith of the Solid State Division is studying the effect of heat treatment at various oxygen pressures on the electrical properties of these crystals. He hopes to demonstrate a relationship between the paramagnetic susceptibility of the excess oxygen and the electrical indications of non-stoichiometry.

Most of the recent work in the Metals and Ceramics Division has already been reported by G. W. Clark and his associates in their Annual Report of May 31, 1963. High-temperature solution growth techniques have produced ThO_2 crystals up to 5 mm, BeO crystals up to 2 mm, and 1-mm crystals of Al_2O_3 , beryl, and SnO_2 . Marginal growth of MgO has been achieved with an arc modified Verneuil process, and surface melting has been achieved with an rf plasma torch. Several Ca-doped KCl crystals have been supplied to the Solid State Division for color center and mechanical property studies, and they have also been supplied a quartz crystal as grown in a KOH solution at 15,000 psi at 400°C for 15 days for electron spin resonance studies. Dendritic-type MgO crystals have also been obtained from a CaF_2 - MgF_2 solution at 1,200 psi at 100°C for several weeks.

The growth of isotopic LiF by C. F. Weaver and his associates in the Reactor Chemistry Division has already been reported in the first bulletin

of the Research Materials Information Center, a recent issue of J. Appl. Phys., and in their Annual Progress Report. Six single crystals of LiF, weighing ~ 300 g each and containing special ^7Li isotopic concentrations ranging from 99.99-79.28% have already been grown. Neutron activation, emission spectroscopy, infra-red transmission, and evidence for strain under polarized light have been used as tools for examination of each crystal. All of these techniques have indicated a continuing improvement in quality. Infra-red transmission has indicated no OH^- contamination, and neutron activation and emission spectroscopy have indicated a total cation impurity concentration of < 10 ppm in the bulk of the more recent crystals. Crystal no. 4, which was 99.99 atom % ^7Li metal, contained only 0.5-1.0 ppm Mn, and no other element was detected. Portions of these crystals are being examined at Cornell University by Prof. R. L. Sproull and Prof. R. O. Pohl to see if they can detect any difference in phonon scattering as a function of phonon wave length between samples of comparable chemical impurity concentration, but of different isotopic concentration.

This work has not proved completely successful to date. The low-temperature thermal conductivity of the isotopic samples has not been significantly better than that of normal isotopic material; hence, they cannot totally differentiate between isotope effects, chemical impurity effects, or such possible unknown effects as that of an unknown impurity, internal strain, U centers, etc. It should be emphasized, however, that these same crystals have already served as improved standards for spectroscopic analysis of LiF and that other portions are being used for neutron scattering, ultra-violet, infra-red, x-ray diffraction, and light scattering experiments.

We have already witnessed one very positive aspect of our efforts to date within the Pure Materials Program at ORNL. Analytical chemists, neutron activation specialists, mass spectrographers, crystal growers, and crystal property researchers have aided themselves and each other by a constant interchange of information as obtained on a particular sample, or on a continuing set of samples. Any improvement in the techniques of one group is therefore reflected in the data and analyses of the other groups. Analytical techniques have therefore been changed, in certain instances,

from the field of routine analysis to that of a continuing program of basic research on a specific material. The presence of unexpected impurities, or the pronounced effect of certain impurities, has sometimes been revealed only by using a wide variety of analytical techniques, procedures in crystal growth, or physical property measurements.

I would now like to say a few words about the extreme importance of careful analyses. Selected samples of high-purity iron were recently prepared by the United States Steel Corporation by purification of ferric chloride and by careful distillation and hydrogen reduction. Identical samples were sent to 7 laboratories, and 5 of these returned their results for a comparative analysis. The results of 3 solids mass spectrometer and 2 neutron activation analyses gave an agreement within a factor of 10 for a large number of elements; however, the results for 11 elements varied greatly and indicated a large variability in analytical techniques, sample inhomogeneity, or both. One can infer from the data that the iron was probably inhomogeneous in Al, Si, and Cu; but one can really tell nothing about the other 8 impurities, including F, Cl, Na, S, K, Ca, Zn, and Ta. The whole point is that each of these 5 analyses differed a great deal from any of the others, yet one cannot hope to use ORNL, U. S. Steel, Bell and Howell, RCA, Westinghouse, Aldermaston (England), and Vitry (France) as a technique for inter-comparison for each individual analysis.

I do not want to be too hard on the analysts. There are books that contain graphs of the electron mobility in Ge as plotted against the year of crystal growth. The minority carrier lifetime in semiconductors was thought to be a sensitive function of something that researchers called "deathnium," until they found that it was copper. A later onset of "deathnium" turned out to be the actual amount of gold that is present in any crystal growth apparatus. Researchers at Bell and Howell have recently doubled the electron mobility in GaAs by reducing the Si content from 1 ppm to 200 ppb.

My point, however, is that any real progress in the production of better crystals will demand a companion progress in improved analytical techniques. We now have tungsten crystals with < 50 ppm total impurity content; however, this laboratory does not have any program or facilities

for isotopic addition techniques to analyze for such impurities as carbon, oxygen or nitrogen in such crystals. The mass spectrometer group of this laboratory was totally committed to "in-house" programs and could not even participate in the round robin analysis of the iron samples from U. S. Steel.

The Pure Materials Program is a continuing program, and it is anticipated that the analytical demands will increase in both quantity and quality. Further purification of starting materials and continued improvement in growth techniques will probably require a parts-per-billion analysis of both starting materials and such end products as metal crystals, alkali halides, intermetallic compounds, oxide semiconductors, and so forth. It would seem that an expansion in the scope of the mass spectrometer analytical services of this laboratory could be justified on the basis of such a program and that careful consideration might be given to the formation of a research-analytical group that would be primarily associated with the problems of those research groups of the laboratory that are concerned with the growth and use of research quality specimens.

It is evident that the success of the Pure Materials Program depends on the continued aid and cooperation of those who analyze, grow, and use research materials. The success of the Research Materials Information Center also depends on the amount of cooperation that is obtained from both crystal growers and users. Background information can be collected from the open literature, but it is woefully inadequate and too late to be of much value. The Center really needs better data sheets (see Fig. 1), internal reports, preprints, and a rapid and personal notification of any new result or development. It is hoped that improved information on improved materials will be of sufficient significant benefit to both producers and users that such cooperation will be continued. It is also hoped that those who analyze, grow, and use research materials will obtain and fill out data sheets of the type indicated in Fig. 1.

Element <u>Compound</u> Alloy	RESEARCH MATERIALS INFORMATION CENTER Solid State Division Oak Ridge National Laboratory P.O. Box X, Oak Ridge, Tennessee		Metal <u>Semiconductor</u> Insulator
Material	AlSb		Assay <u>99.9999</u> %
Form (single crystal, whisker, rod, etc.)	single crystal		Impurity (dopant or isotopic enrichment) ppm
Dimensions	1-inch diameter - up to 4 inches		S 1 to 100
Orientation	Long		Se 1 to 100
			Te 1 to 100
			Cd 1 to 100
			Zn 1 to 100
Starting Material (and purity)	Al	99.9999%	
	Sb	99.9999%	
Method of Production (reference pertinent report or paper where possible)	Czochralski. W. P. Allred, "Crystal Growth of III-V Compounds" in <u>Ultrapurification of Semiconductor Materials</u> , edited by M. S. Brooks and J. K. Kennedy (The Macmillan Company, New York, 1962), p. 550.		
Method of Final Analysis	Mass spectrograph. R. Brown, R.D. Craig and J.D. Waldron, "Analysis of Impurities by Spark Source Mass Spectrometry," in <u>Preparation of III-V Compounds</u> , edited by R.K. Willardson and H.L. Goering (Reinhold Publishing Corporation, New York, 1962); Electrical. F.J. Reid, <u>Electrical Analysis of III-V Compound Semiconductors</u> , "Ibid., Residual impurities 1.0 ppm, p-type and 0.5 ppm, n-type.		
Research Interest or Applications	Diodes, Solar Cells		
Special Characteristics (include handling precautions)	Hole mobility at 77°K 800 cm ² /volt sec 1300 2000 3500		Carrier Concentration 1 x 10 ¹⁷ /cm ³ 3 x 10 ¹⁶ 1 x 10 ¹⁶ 3 x 10 ¹⁵
Availability for External Distribution	\$10/gram		delivery: 3 weeks
Name of Producer	Bell & Howell Research Center		
Installation	360 Sierra Madre Villa		
Address	Pasadena, California		
	TX-3309 (6-62)		

Fig. 1. Research materials data sheet.

OLD ARMOR PLATE AND LEAD

P. S. Baker, ORNL

At one time or another each one of us runs across a material -- or perhaps a service -- which is out of the ordinary and which, for that reason, turns out to be of particular interest or value in our work. I should like to mention a few of these which have crossed our paths.

Old Armor Plate

There appears to be a sizeable quantity of armor plate from old battleships which can be made available rather cheaply and which is quite suitable for shielding purposes. Information can be obtained from

Procurement Branch
Office of Controller
U. S. Atomic Energy Commission
Washington, D. C.

Old Lead

In Holland there is a batch of about 75 tons of lead which has recently been raised from a ship sunk in 1806. This is, of course, fallout-free and may be of interest to some people. Questions concerning this source of supply should be directed to

Dr. J. A. Goedkoop
Reactor Technology Laboratory
Patten, The Netherlands

At ORNL we are continually looking for unusual isotopic distribution in elements used in our electromagnetic separations programs. During the past five years we have run across two sources of lead with atypical isotopic abundances. The first batch occurred in tailings from thorium-containing ores and was recovered in the rare-earth stream. It was processed by the Heavy Minerals Company in Chattanooga, Tennessee, and we obtained about 10 Kg of material. Below is shown the isotopic ratio as compared with what we usually call "natural lead." (The term "radiogenic" is used rather loosely here, since I presume that all lead could be considered to be of radiogenic origin.)

LEAD "208"

Isotope	Abundance, %	
	Natural	Radiogenic
-204	1.48	0.04
-206	23.6	25.6
-207	22.6	1.8
-208	52.3	72.6

In 1961 we ran across several sources of rather large quantities of lead which were high in ^{206}Pb . Industrial Metals Improvement Company in Pittsburgh had 18 tons of lead oxide from Canadian pitchblende which was about 80% ^{206}Pb and contained, in addition to the usual 204, 207, and 208, traces of 203, 205, 209, and 210.

At about the same time, National Lead Company at Perth Amboy, N. J., told us that they had nearly 100 tons of lead recovered during the refining of uranium ores from the Congo. Some was as PbCO_3 and some as PbO and, interestingly enough, they had different assays:

LEAD "206"

Isotope	Abundance, %	
	PbCO_3	PbO
-204	< 0.05	0.06
-206	92.63	88.98
-207	6.38	8.25
-208	1.00	2.71

Stable Fission Products

A third item which I want to include, mentioned by A. F. Rupp (page 81), is the recovery of stable isotopes from fission products. We have separated a quantity of neodymium, which has an "abnatural" (a new word analogous to "abnormal") isotopic distribution. The big differences are in lack of 142 and higher ratios of 143 and 145.

NATURAL NEODYMIUM	
<u>Isotope</u>	<u>Abundance, %</u>
-142	27.7
-143	12.4
-144	23.8
-145	8.3
-146	16.8
-148	5.6
-150	5.4

FISSION NEODYMIUM	
<u>Isotope</u>	<u>Abundance, %</u>
-143	27.7
-144	26.5
-145	19.1
-146	14.9
-148	8.3
-150	3.5

ORNL Target Center

E. H. Kobisk, ORNL

The ORNL Target Center was commissioned by the AEC to produce targets of stable and radioactive isotopes for nuclear research purposes. These targets were intended primarily for use in linear accelerators, cyclotrons and reactors to study charged particle scattering characteristics, nuclear reactions, and to determine reaction cross sections (including fission cross sections). Therefore, most targets assume the form of thin films of isotopically enriched elements (or their compounds) on a backing (usually copper, stainless steel, or tantalum) or in self-supporting form. Most accelerator targets of the thin film type have areal densities between $5 \mu\text{g}/\text{cm}^2$ to $10 \text{mg}/\text{cm}^2$.

When the Target Center was formally organized in 1961, the total production of targets was about 35 per year. At present the fabrication level has increased to more than 100 per month. Initially the fabrication group consisted of four technicians and four technical personnel; now the personnel complement consists of three technicians and four technical men. The vastly increased production level is being maintained and further enhanced by the addition of new and more effective equipment as well as by greatly improved technology, most of the technology being developed by our own personnel. Distribution of these targets is made on an international basis, each target being prepared to order and specification of the researcher. Under this arrangement, targets have been supplied by the Center to more than 150 colleges and universities and to most of the National Laboratories. In addition, targets have been prepared for use in more than 20 foreign countries including Australia and South Africa.

Many techniques are used to prepare targets, but the most important of these are:

1. Vacuum evaporation and condensation of isotopes into thin film form.
2. Rolling of isotope metals into very thin foils having areal densities between $0.5 \mu\text{g}/\text{cm}^2$ and $300 \text{mg}/\text{cm}^2$.
3. Pressing and sintering of heavy samples (powder metallurgy).
4. Electrodeposition of isotopic metals from aqueous solution.

Using these techniques, targets have been prepared of each element in the periodic table excluding the inert gases and a few of the man-made, radioactive elements. During the last year, targets of radioactive elements have been successfully prepared. Included were such materials as most of the plutonium isotopes, uranium isotopes, americium, radium, neptunium, etc. These latter isotopes were all fabricated into thin films on metal backings or as thin films on thin carbon backings which could be used as self-supporting targets in the tandem Van de Graaff accelerator.

Because vacuum evaporation of materials requires very high temperatures, sometimes in excess of 3000°C , and because this technique is generally inefficient in terms of isotope utilization, most developmental efforts have been directed at improving this particular technique. Using electron bombardment -- similar to the electron kinetics of a radio vacuum tube -- temperatures in excess of 3000°C have been attained. Development of crucibles to hold milligram quantities of isotopes and to direct the beam of vaporized material has been successfully made which can, under appropriate conditions, increase the isotope utilization efficiency to over 90%. Compared with the usual efficiency of 1% obtained with resistance filaments, this represents considerable conservation of isotope utilization. The result of this development is reduced isotope cost to the customer desiring a target. Also, this technique has provided a method for evaporation-condensation of extremely costly isotopes previously not attempted because of the prohibitive cost to the customer for isotopic starting material.

In the future, emphasis will be placed on the further development of techniques to reduce isotope cost and to improve target uniformity. During fiscal years 1964 and 1965, a facility is to be constructed in which the rolling of thin foils of plutonium, neptunium, uranium, thorium and americium can be done. Also in this facility, targets of almost all alpha-active materials will be possible to fabricate as well as many other gamma- or beta-active isotopes. Presently some of these foils and targets are being prepared on a small scale limited by the containment facilities and equipment available.

Another important future advancement will be the preparation of single crystals and hyper-pure isotopes using zone refining and other appropriate techniques. Recently some success was attained in forming thin films (500 A. thick) of silver metal having single crystal orientation of 1,0,0. More effort in this area is anticipated. The desire of researchers to obtain highly precise targets is indicated by the organization of the Euratom Laboratories (EANDC) at Mol-Geel, Belgium. This laboratory has been directed by Euratom to prepare standard targets to be used for the measurement of neutron cross sections and other physical information needed to promote the expanding reactor programs within the European Community.

During the author's visit to these Euratom laboratories, August, 1963, detailed observations were made of their target fabrication facilities and techniques. In summary, these observations together with many interviews of personnel employed at CBNM (Central Bureau of Nuclear Measurements) led the author to believe that the primary function of their services was to furnish highly precise targets of fissile isotopes and a few stable isotopes for cross-section determinations, reactor oscillation measurements, etc. One example of this function was the preparation of three sets of boron targets: elemental

boron deposited on quartz plates, quartz tubes, and boron (borate) in water solution. These samples were used for oscillation measurements and all such measurements agreed to $\pm 0.3\%$. This precision could have only been achieved by definition of thickness, uniformity, and purity to $\pm 0.1\%$ in each sample. Although such high precision work is being done at CBNM, it is well to note that these boron samples took more than two years to prepare.

PREPARATION OF SPECIAL ISOTOPES AND
OTHER SERVICES AT BROOKHAVEN

L. G. Stang, Jr., BNL

I am grateful for the opportunity to discuss some of the work being carried on at Brookhaven National Laboratory. I shall confine my remarks to work being performed within the Hot Laboratory Division of the Nuclear Engineering Department and more particularly to that part of the work which is of interest to groups outside of Brookhaven. Thus, I will restrict my comments to the development and production of what we term "special" isotopes. Such isotopes are mostly short-lived, although some are long-lived and some are even stable. Their applications lie largely in the field of medicine, although some are used industrially. A feature in common is that all require chemical processing following irradiation. Another chief feature is that none is supplied commercially by any other source of supply in the United States (at least not in the form supplied by Brookhaven). The latter is important because we look upon our work as supplementing other sources of supply in this country rather than competing with them.

Routine Production

The following isotopes are produced routinely for sale outside of Brookhaven. By "routine production" we do not imply that each isotope is necessarily produced every week or according to a pre-arranged fixed frequency, but rather that each of the following products is produced according to a fixed procedure as orders are received without any further development being required.

Reactor

- ^{18}F : 1.7 hr, pure positron emitter, the only practical radioactive tracer for fluorine, produced by neutron irradiation of Li_2CO_3 , 8 mc units (larger amounts possible).
- ^{28}Mg : 21.3 hr, beta emitter, the only practical radioactive tracer for magnesium available, specific activity 40 mc/g, made by neutron irradiation of $^6\text{Li}^{26}\text{Mg}$ alloy.
- ^{132}I Generator: 2.3 hr (77-hr parent ^{132}Te), fission product, carrier free. A generator is a device by which a short-

lived daughter can be milked simply, conveniently, rapidly, and repeatedly from a longer-lived parent. Some of the advantages of any and all of our generators are:

1. Carrier-free products
2. Combine the advantages of shipping a long-lived isotope with the advantages of using a short-lived one, to wit:
 - a. Lower radiation dose to patient
 - b. Repeated measurements readily made on the same system without having to correct for residual radioactivity present from previous measurement
 - c. Waste disposal eliminated
3. Convenient operation
 - a. Rapid
 - b. Simple
 - c. Repeated milkings possible

See references listed at the end of this talk for more complete discussion of generators in general and of other details of these various isotopes.

^{99m}Tc Generator: 6.0 hr, pure-soft-gamma-emitter (140 Kev), (67-hr parent ^{99}Mo), daughter ^{99m}Tc is virtually stable, carrier free.

^{90}Y Generator: 65 hr, (28-year parent ^{90}Sr), ^{90}Sr contamination activity is 10^{-8} times ^{90}Y activity, both ^{90}Y and parent ^{90}Sr activity are pure-beta-emitters, carrier free.

^{28}Al Generator: 2.28 min, (21-hr parent ^{28}Mg), carrier free.

^{132}Te : 77 hr, available in solution form.

^{99}Mo : 67 hr, available in solution form.

^{133}I : 21 hr, carrier free, fission product, maximum radiopurity of 80% occurs at 30-50 hr following irradiation (^{132}I , ^{135}I , and ^{131}I also present).

^{99m}Tc Colloid: Particle size $\sim 100 \mu$, ideal for scintillation scanning applications (low dose to patient, high contrast on film, good detail, blood clearance half-time of ~ 3 min), activity is firmly fixed to colloidal sulfur.

^{90}Y Beads: Fused clay matrix (physiologically inert), variety of sizes with or without attachment to implanting wire,

1-2 mc/bead (as desired), can provide complete destruction of tissue within ^{90}Y beta range.

- ^{47}Sc : 3.4 day, carrier free, produced from ^{47}Ca decay.
- ^{38}Ar : Stable, exceedingly high purity, 0.03 cc units (STP), from neutron irradiation of NaCl, used for calibrating mass spectrographs.
- ^{128}Xe : Stable, exceedingly high purity, 0.03 cc units (STP), from neutron irradiation of NaI, used for calibrating mass spectrographs.

Accelerator

- ^{67}Cu : 61.6 hr, carrier free, enriched $^{64}\text{Ni}(\alpha, p)^{67}\text{Cu}$, used in studies of Wilson's disease.
- ^{43}K : 22 hr, carrier free, $^{40}\text{Ar}(\alpha, p)^{43}\text{K}$.
- $^{87\text{m}}\text{Sr}$ Generator: 2.8 hr (80-hr ^{90}Y parent), carrier free, pure medium energy (390 Kev) gamma, $(\alpha, 2n)$ on natural RbCl.
- Protein Iodinated with ^{124}I : 4.5 days, positron emitter, (α, n) on natural antimony.
- ^{68}Ga Generator: 68 min (250-day parent ^{68}Ge purchased from ORNL), carrier free, positron emitter.

Currently Being Developed

It must be emphasized that the items in this section are still in the developmental stage and are not yet ready for routine production. This list is merely representative and does not include all items currently under development in the Hot Laboratory Division of Brookhaven.

At Others' Requests

- ^{52}Fe : mc amounts, via enriched $^{50}\text{Cr}(\alpha, 2n)^{52}\text{Fe}$.
- ^{132}Cs : Via $^{129}\text{I}(\alpha, n)^{132}\text{Cs}$ using a thallium iodide target.

On Our Own Initiative

- ^{28}Mg : High specific activity via recoil techniques.
- $^{99\text{m}}\text{Tc}$ colloid of different particle sizes to localize at different sites.

Isomer generators: ^{127g}Te , ^{129g}Te , ^{131g}Te to be milked from corresponding metastable state parents.

^{42}K Generator: 12.5 hr (> 3.5 -year ^{42}Ar parent), carrier free.

Study of selenium chemistry for possible development of ^{72}As generator.

Triton-induced reactions in general, especially those occurring with reactor-produced tritons.

^{48}Sc : 44 hr, carrier free, via $^{51}\text{V}(14\text{-Mev } n, \alpha)$.

^{54}Mn : 300 days, high specific activity, from stainless steel to be irradiated parasitically at high flux [$^{54}\text{Fe}(n, p)^{54}\text{Mn}$].

References

The following two references may be of general interest and will also provide additional specific details:

1. A Review of the Production of "Special" Radioisotopes, L. G. Stang, Jr., presented at the Seminar on the Practical Applications of Short-Lived Radioisotopes Produced in Small Research Reactors held in Vienna, November 5-9, 1962, under the sponsorship of the International Atomic Energy Agency.
2. Tailoring the Isotope to the Need, L. G. Stang, Jr. and P. Richards, Nucleonics, January 1964.

CYCLOTRON SERVICES

J. E. Beaver, ORNL

The 86-Inch Cyclotron is a fixed-frequency proton accelerator, operating at a frequency of 13.4 Mc and with a field strength of $\sim 9,000$ gauss. It accelerates protons to an energy of 22 Mev. The cyclotron is available for service irradiations (including the production of neutron-deficient radioisotopes, basic physics research, and bombardment of electronic components and biological specimens) for approximately 80 hr per week.

Cyclotron-produced radioisotopes have, in general, high specific activities, since they are ordinarily of a different chemical element than the target and hence are quite frequently carrier free. The cyclotron-produced isotopes usually lie on the neutron-deficient side of the nuclear stability line, and their principal mode of decay is by positron emission and electron capture. The relatively high production rates on the 86-Inch Cyclotron give a rather inexpensive source of cyclotron-produced isotopes. For example, the production rate for the 2.7-year half-life ^{55}Fe is 8 mc/hr (for a 10-hr bombardment, the cost per mc including all incidental charges would be $\sim \$12/\text{mc}$), and for the 270-day half-life ^{57}Co , the production rate is 17 mc/hr.

Radioisotope production is usually performed with the internal beam, where 1 of 3 basic target types is used: flat plates, capsules, and window targets. Flat-plate targets consisting of a 6-in. x 5-5/8-in. x 1/2-in. base plate, usually of copper, are used where the actual target material, such as Li, Pb, Ni, Ta, Ga, can be readily bonded to the base. Fig. 1 shows a typical Ni-plated copper flat plate. About twenty-one 3/16-in. water passages drilled longitudinally through the base provide the cooling required to dissipate the high power input into the target. Operation at 1 ma requires that 22 kw be dissipated. The target is installed in the cyclotron in a horizontal plane tangent to the beam at a radius of ~ 31 in. The beam will strike the leading edge of the target area at about a 6° angle and will then be spread over an area $\sim 1\text{-}1/2$ in. wide x $\sim 3\text{-}1/2$ in. long. The energy of the protons striking a flat-plate target can be varied by distorting the magnetic field with auxiliary coils on the pole pieces of the main magnetic, which results in decreasing the radius of the beam. A minimum energy of ~ 17.5 Mev with intensities up to 2,500 μa

PHOTO 37959

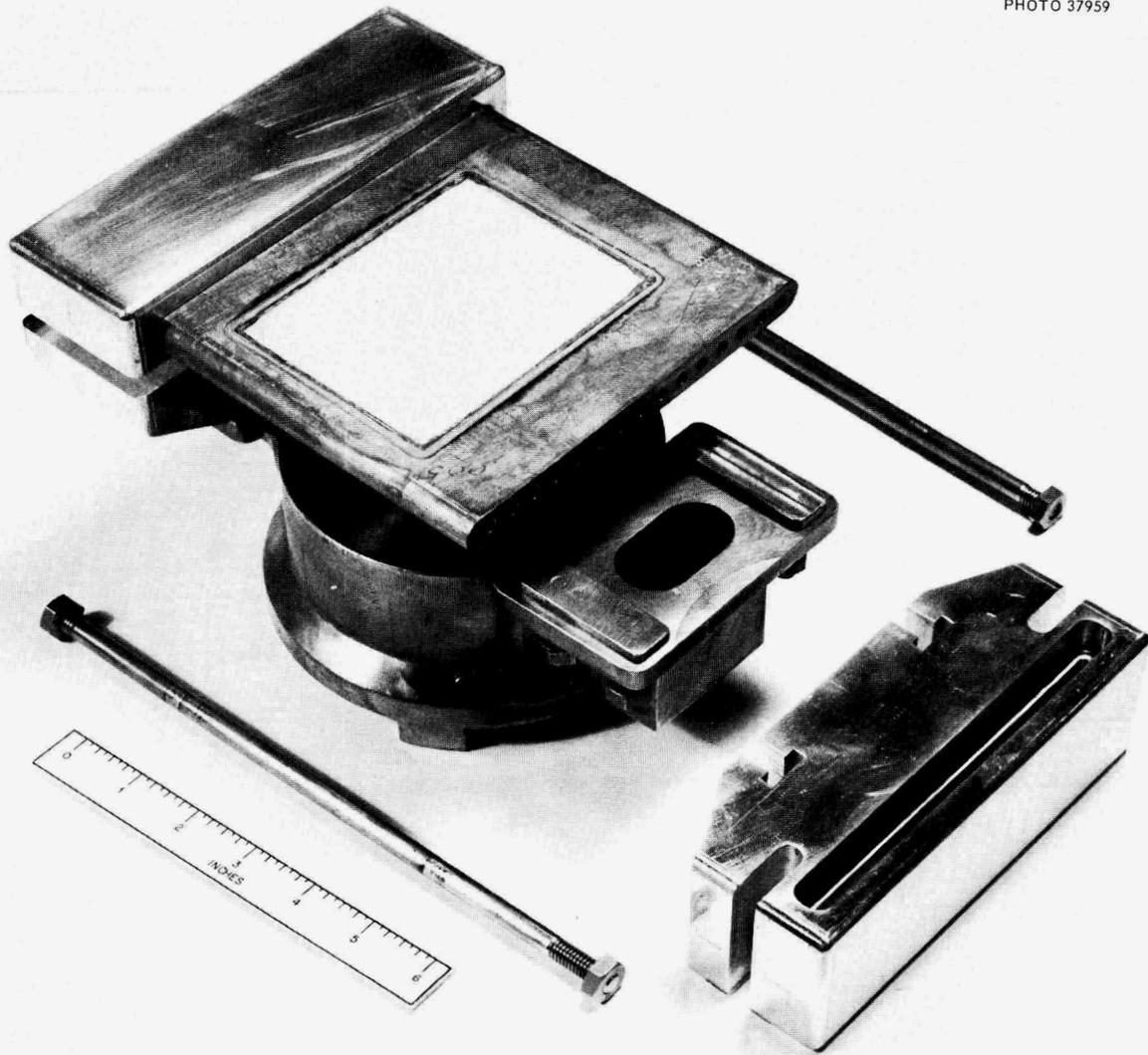


Fig. 1. High-Current Flat-Plate Target Partially Assembled in Header Unit.

is available. The maximum energy for flat-plate targets is ~ 21 Mev with intensities up to $1,500 \mu\text{a}$. The beam current at which a target can be bombarded depends on the melting and vaporization point of the target material. Flat-plate targets are used for (p,n) , $(p,2n)$, and (p,α) reactions, with the $(p,2n)$ reaction having the highest cross section.

The standard capsule targets, like the one shown at the bottom of Fig. 2, are 10 mil wall, 200 mil OD, Ni or Al water-cooled tubes $\sim 5\text{-}1/2$ in. long. A 3-in. cavity in the center of these tubes provides a means of irradiating chemical compounds or metal powders at up to $180 \mu\text{a}$ beam current. The capsule is inserted into a $1/4$ -in. dia jacket shown just above the capsule. About 7 gal of water per min, at a pressure of 120 lb, flows in a 15-mil annular ring between the capsule and the water jacket. The middle target in Fig. 2, with the outer jacket shown just above it, is an experimental target in which it is hoped to increase the beam current by about 50% by increasing the amount of cooling water around the capsule. The upper target is a modified tube target made by flattening a $1/2$ -in. Cu tube in the center and electroplating high-purity Au onto the flattened surface. Capsule targets are used where coating on a flat plate is extremely difficult or where the production is more economical by using the less expensive capsule. Capsule targets are used for the (p,n) reaction rather than the $(p,2n)$ reaction because the beam energy is reduced several Mev after passing through the outer water jacket, the 15 mils of cooling water, and the capsule.

Fig. 3 shows a standard window target. The beam will strike this target at the center, almost perpendicular to the milled recess. Window targets are used where the amount of target material is limited, as in the case of electromagnetically enriched isotopes. The target material, normally $\sim 30\text{-}40$ mg, is usually wrapped with 1 or 2 layers of 2-mil Al foil and placed behind the 12-mil Al window. Fig. 4 shows the foil wrap and the inside of the window. The bronze clips are used to hold the foil in place against the window. Because of the inefficient cooling in the window target, the beam intensity is limited to $\sim 80 \mu\text{a}$. Window targets are used for $(p,2n)$ reactions and are used with appropriate absorbers for (p,n) and (p,α) reactions.

PHOTO 36590

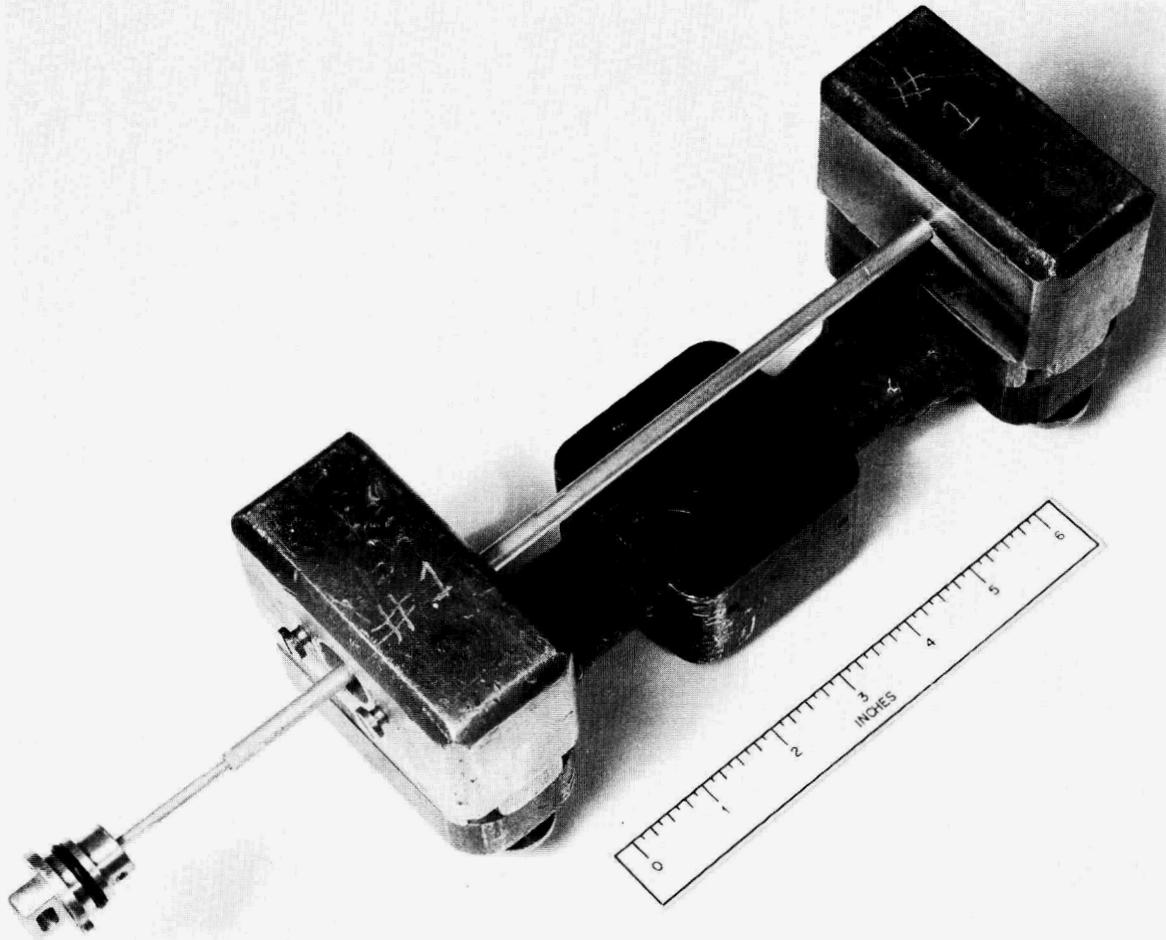


Fig. 2. Capsule Target Partially Assembled.

PHOTO 36593

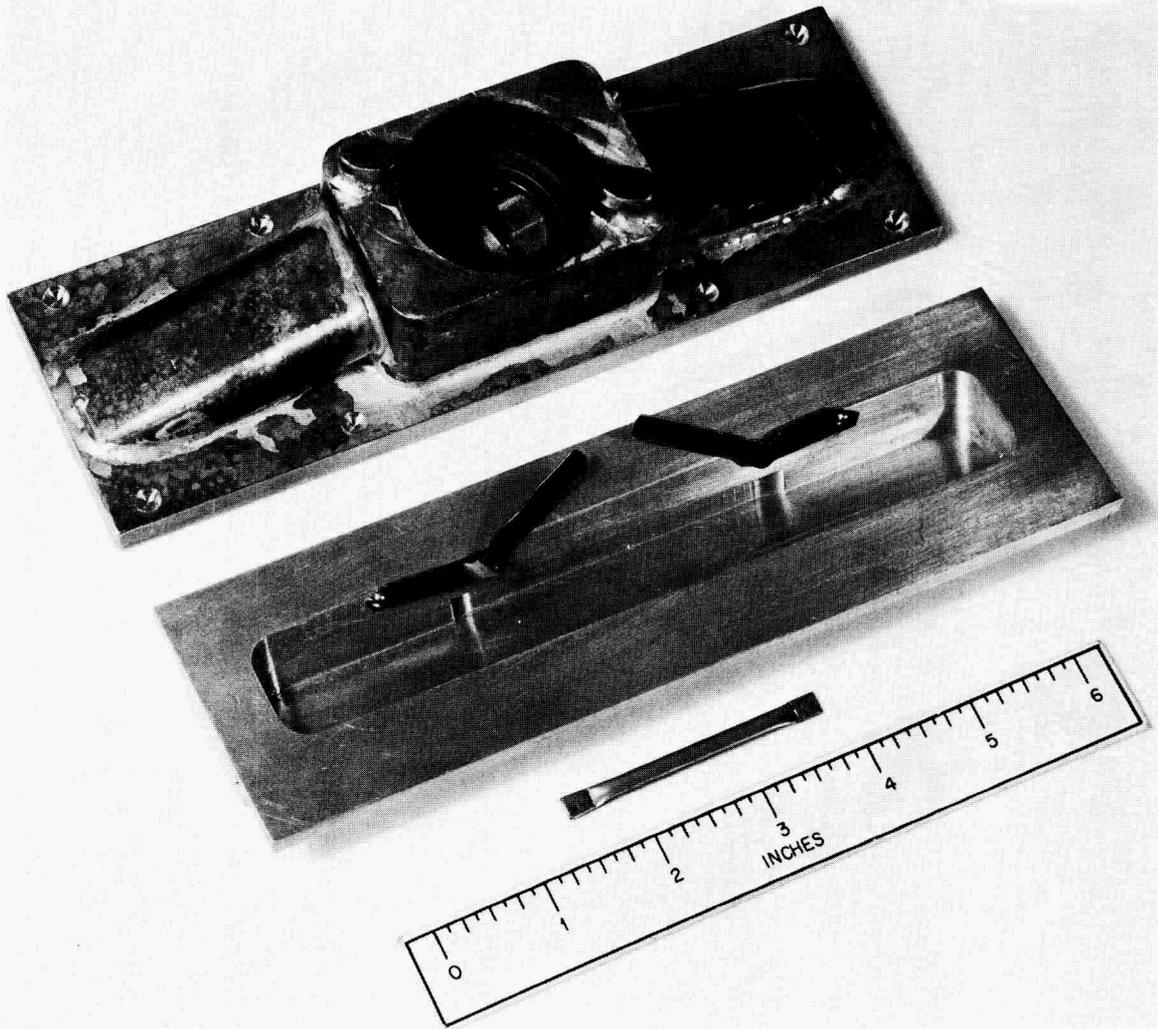


Fig. 3. Window Target. The filled and sealed tube in the foreground is held in the slotted window area by the bronze clamps.

The deflected beam is deflected from the main magnetic field by means of a magnetic channel coil which directs the beam into a 2-in. beam pipe ~ 50 ft long. The deflected beam is used primarily for experimental work rather than radioisotope production, with the exception of the bombardment of Kr gas to produce ^{84}Rb .

There are 3 irradiation ports located in the beam pipe. They are referred to as the "T position," "0° port," and "15° port." At the "T position" (located in the cyclotron pit ~ 15 ft from the dees), beam intensities up to 15 μa are available. The beam is slightly elliptical in cross section, with the major axis being about an inch long. The beam is not homogeneous, the intensity at the center being an order of magnitude greater than the outer edge. Due to limited space, targets at the "T position" normally consist of foil stacks, or powders packed between 2 foils with the beam passing through a 5/8-in. collimator. Several water-cooled probes are available for "T position" bombardments.

At the "0° port," which is located at the end of the beam pipe outside the cyclotron pit, beam intensities between 10^{-6} μa and 10^{-1} μa are available. For experimental work, the "0° port" is more versatile than the "T position." Target probes are available for bombardments at atmospheric pressure, at the cyclotron vacuum, or in an inert atmosphere. The "0° port" is used for such bombardments as foil stacks, electronics components (such as transistors, solar cells, and diodes), and biological specimens.

CALUTRON SERVICES

W. A. Bell, ORNL

The word "services" used in connection with our calutron work is a misnomer because most projects undertaken have value both to us and to the requester. Thus, in place of "services", I will talk more in terms of special projects which are performed through the use of calutron equipment. It should be pointed out that these projects are not performed daily; instead they are fitted into the program at opportune, irregular intervals.

The calutron is useful in completing numerous projects not directly related to replenishing the inventory of separated isotopes. Perhaps the most important types of special projects which we perform are as follows:

- a. Target preparation (in limited extent).
- b. High-purity isotope separation.
- c. Low-activity radioisotope separation.
- d. Chemical purification by means of mass separation.
- e. Research activities of our own initiation.

Let me cite some specific examples.

In the field of target preparation, one utilizes the principle of ion deposition in or on a suitable substrate. Any ion species can be placed on almost any substrate in quantities of 10-20 $\mu\text{g}/\text{cm}^2$ by direct use of the calutron. This includes both gaseous and solid materials. The maximum amount of a solid which can be deposited from a 40-kv ion beam is dependent upon the ion and the substrate, but generally lies within the limits of 10-100 $\mu\text{g}/\text{cm}^2$. This limitation is imposed by the sputtering process encountered whenever a 40-kv ion strikes a solid surface. In general, several atoms (5-8) of target substrate are removed by each ion impingement. Sputtering is known to decrease rapidly as ion energy approaches 100 volts; hence, ion deceleration is sometimes employed in target formation studies. We find that whenever ions are decelerated to the 100-volt region, deposits of 250-1,000 $\mu\text{g}/\text{cm}^2$ can be obtained, but difficulty is sometimes encountered in acquiring uniformity and adherence of the deposit. Specific examples of this work using 40-kv ion deposition include:

- a. N₂ or Ar on Ta, yielding 20 μg/cm².
- b. Pb on Ni, yielding 10 μg/cm².
- c. Th on C-18 type graphite, yielding 65 μg/cm².
- d. Th on pyrolytic graphite, yielding 30 μg/cm².

Examples of deposits achieved utilizing the deceleration technique include:

- a. B on Ta, yielding 250 μg/cm².
- b. Mg on Cu, yielding 1,000 μg/cm².

High isotopic purity relative to the initial abundance found in nature is achieved through development of specific equipment, alteration of operating techniques, or performance of second-pass separations utilizing pre-enriched feeds. Development and construction of the 255°, 0.5 inhomogeneity separator placed within the calutron system makes possible the achievement of higher enrichments from a single-pass process. Two of these separators are now in constant use. They provide ion currents equal to those in the calutron with the following typical increases in isotopic purity over the best calutron-separated samples:

1. ¹⁰⁸Cd from 71-86.5%.
2. ¹¹⁷Sn from 90-93%.
3. ¹⁸⁰Ta from 0.28-0.45%.
4. ²⁰⁴Pb from 76-86%.
5. ²⁰⁶Pb from 81-98%.

Second-pass separations have been completed to acquire isotopic samples of purity ranging from 99.9-99.999%. Isotopes included in this purity range are ¹⁰B, ⁴⁰Ca, ⁵²Cr, ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁸Pb, ⁶Li, ¹⁶⁶Er, ¹⁶⁸Er, ¹⁵⁶Gd, ¹⁴⁸Gd, ¹⁵⁰Sm, and others.

Low-activity radioisotope separations (except U, Th, and Pu) have been limited to feeds of irradiated materials such as ¹⁰Be, ¹⁰⁸Ag, ²⁰⁸Bi, and ²¹⁰Bi. In each instance the active isotope existed only in the ppm range. A ¹⁰Be separation was made from feed having 2 ppm of the desired isotope. From this we achieved an 8-mg sample of Be containing 0.29% ¹⁰Be - an enrichment factor of 1370. It is interesting to note that here the ¹⁰Be had penetrated to a depth of 1/8 in. into the graphite collector pocket.

The ¹⁰⁸Ag separation was made from feed containing only 1 part ¹⁰⁸Ag in 40 x 10⁶ parts of total Ag. The sample attained was enriched by a

factor of 55, and all traces of ^{110}Ag were removed. The enrichment was less than was predicted from calutron parameters and this was disappointing, even though the sample was still of useful quality. The separation of ^{208}Bi and ^{210}Bi was attempted twice. The first separation was a miserable failure, yielding an infinitesimally small sample enriched by a factor of only 2. The next attempt, 2 years later, redeemed us to a certain extent, but we still think it should have been better. Here 90 g of Bi, containing 25 ppm ^{208}Bi and 150 ppm ^{210}Bi , was processed as part of the shakedown program required to test separators prior to starting Pu separations in the new containment facility.

Chemical purification by mass separation was first performed by the calutron group in 1953. At that time pure Tb was difficult to obtain, and a few mg were separated from other rare earths during a routine processing of Gd. The Gd was in no way compromised by this approach, and a sample of Tb was produced which was free of all rare-earth contaminants except Gd, which appeared in small proportion. In a similar manner yttrium was processed through the calutron to remove all rare-earth contaminants. In this work yttrium chloride was used as the charge material, and it was predicted that minute contamination by a complex chloride ion of dysprosium was possible. The yttrium sample was freed of rare earths down to the ppb range, and the only contaminating rare earth detected was the dysprosium which entered the collector pocket as a complex ion.

Other special projects, either requested or initiated by ourselves, include:

1. Studies in plasma physics involving ion beam interactions and beam stabilization.
2. Studies pertaining to ion retention by specifically oriented single crystals.

In the latter work we grow our own crystals and bombard them with ^{46}Ca ions. The highest ^{46}Ca purity, 51% from 0.0033%, was achieved during collection of the ions on a copper crystal having the (110) face lattice perpendicular to the incoming ion beam. Ion retention by single crystals is of sufficient academic interest that one of our men is using experiments in this field as project work for his master of science degree.

In summary, it is evident that the Isotopes Division has profited technically from performing these projects. This technical advancement,

in addition to the current availability of numerous calutrons supported by a pool of technically trained operating supervision, makes an increase in the number and variety of "service" projects undertaken not only possible but desirable. We invite your interest in utilizing the calutrons whenever they can be of help to your program.

PRODUCTION QUANTITIES OF ISOTOPIES BY GASEOUS DIFFUSION,
MOLECULAR DISTILLATION, AND CHEMICAL EXCHANGE

S. A. Levin, ORGDP

The isotope production groups at Oak Ridge have received many requests for information regarding the feasibility and costs of producing enriched isotopes which are of interest in various phases of the Government's nuclear program. Our studies have indicated that isotope separation facilities which are available in our production plants can be used for the production of certain of these isotopes, in particular the isotopes of tungsten, molybdenum, xenon, chlorine, and lithium at costs which are attractive.

The facilities which are available at present consist of gaseous diffusion stages and the lithium isotope separating facilities. In addition, other isotope separation facilities could be constructed at relatively low cost because of the skills and facilities which already exist here. Specific facilities which have been studied in view of anticipated requirements are: chemical exchange columns for the production of boron-10 and molecular distillation trays for the production of potassium-39.

In my talk I will discuss isotope separation by gaseous diffusion, chemical exchange, and molecular distillation.

Gaseous Diffusion

There is at the present time a group of stages at the Oak Ridge Gaseous Diffusion Plant which is not being utilized for the production of U-235. Additional stages may be made available in the future. Specific studies performed so far indicate that the elements tungsten, molybdenum, xenon, and chlorine have potentially useful isotopes which may be obtained at minimum costs by the gaseous diffusion process. The cost estimates which will be presented for isotope production by means of existing gaseous diffusion equipment represent out-of-pocket costs to the Government which would be prorated to the particular isotope production program. No charge for the depreciation of existing equipment was included.

Tungsten-184

Tungsten, because of its extremely high melting point (3400°C) is the most attractive metal for use in fabricating fuel elements for nuclear rocket engines. Naturally occurring tungsten has a thermal neutron absorption cross section of 17.5 barns as is shown in the first table. Engine weights can be reduced and the structural integrity of the fuel elements can be greatly increased by reducing the absorption cross section. To do so, it is required to enrich the low cross section isotope W-184 from a mixture of five isotopes.

We prepared estimates of the costs of producing material with an absorption cross section of from 3 to 5 barns. Tungsten with a cross section of 3.5 barns can be produced at a rate of 8,000 lbs per year at a steady-state unit

Table 1.

TUNGSTEN ISOTOPE SEPARATION

Isotope	Cross Section (barns)	Natural Abundance (atom %) $\sigma = 17.5$ Barns	Product Composition (atom %)	
			$\sigma = 4.4$ Barns	$\sigma = 3.5$ Barns
W-180	60	0.14	0.62×10^{-4}	0.13×10^{-4}
W-182	20	26.46	2.40	1.10
W-183	11	14.40	13.00	8.70
W-184	2	30.60	82.20	88.50
W-186	35	28.40	2.40	1.70
Production Rate			12,000 lbs./yr.	8,000 lbs./yr.
Unit Cost			\$850/lb.	\$1300/lb.

cost of \$1300 per lb. Tungsten with a cross section of 4.4 barns can be produced at a rate of 12,000 lbs per year at a steady-state unit cost of \$850 per lb.

Molybdenum-98, 100 or Molybdenum-92, 94

There is interest in low cross section molybdenum for use as a membrane between the core and blanket in a molten salt breeder reactor. Naturally occurring molybdenum has a thermal neutron absorption cross section of 2.7 barns as shown in the next slide. Certain design improvements may be achieved by lowering the cross section. This may be done by enriching the end isotopes, Mo-92, 94 from a mixture of seven isotopes as shown. Similar reductions in cross section can be achieved by enriching the other end isotopes Mo-98, 100.

We prepared estimates of the cost of producing molybdenum with a reduced absorption cross section. Molybdenum with a 0.4-barn cross section can be produced at a rate of 8,000 lbs per year at a steady-state unit cost of \$500 per lb. Molybdenum with a 1.3-barn cross section can be produced at a rate of 25,000 lbs per year at a steady-state unit cost of \$150 per lb.

The first two tables are indicative of the type of multicomponent separation problems one is confronted with in the production of isotopes. Table 3 summarizes the results of our studies for gaseous diffusion and for chemical exchange and molecular distillation as well. We have discussed already the separation of tungsten and molybdenum isotopes.

Table 2.

MOLYBDENUM ISOTOPE SEPARATION

Isotope	Cross Section (barns)	Natural Abundance (atom %)		
		$\sigma = 2.67$ Barns	$\sigma = 1.3$ Barns	$\sigma = 0.4$ Barns
Mo-92	0.3	15.86	76.30	96.00
Mo-94	0.0	9.12	13.70	3.00
Mo-95	13.4	15.70	7.73	0.85
Mo-96	1.2	16.50	1.98	0.14
Mo-97	2.1	9.45	0.23	0.01
Mo-98	0.45	23.75		
Mo-100	0.20	9.62	0.06	0.002
Production Rate			25,000 lbs./yr.	8,000 lbs./yr.
Unit Cost			\$150/lb.	\$500/lb.

Table 3.

PRODUCTION QUANTITIES OF SEPARATED ISOTOPES

Desired Isotope	Product Concentration or Neutron Absorption Cross Section	Production Rate	Unit Cost
<u>Gaseous Diffusion</u>			
W-184	3.5 barns	8,000 lbs. W/yr.	\$1300/lb.
	4.4 barns	12,000 lbs. W/yr.	\$850/lb.
Mo-92, 94	1.3 barns	25,000 lbs. Mo/yr.	\$150/lb.
	0.4 barns	8,000 lbs. Mo/yr.	\$500/lb.
Xe-134, 136	1.0 barns		\$450/kg.
Cl-37	95-99%	17,000 lbs. Cl/yr.	\$150/lb.
<u>Lithium Separation</u>			
Li-7	99.99% (0.13 barns)		\$120/kg.
<u>Chemical Exchange</u>			
B-10	95%	1,800 kg. B/yr.	\$450/kg.
<u>Molecular Distillation</u>			
K-39	99.93%	2,000 lbs. K/yr.	\$560/lb.

Xenon-134, 136

The xenon isotopes of interest occur in the mixture of xenon isotopes obtained as an off-gas from reactors. The xenon separation problem is that of removing the light isotopes of xenon from the mixture so that the resulting material will be suitable for use in certain types of radiation monitoring devices.

We prepared estimates of the cost of producing xenon with a one-barn cross section. Kilogram quantities of xenon per year can be produced at a steady-state unit cost of \$450 per kg.

Chlorine-37

Naturally occurring chlorine consists of 75% Cl-35 and 25% Cl-37. Chlorine isotope separation is desirable in order to remove Cl-35 because of its severe (n,p) cross section for fast neutrons. Chlorine enriched in Cl-37 to a concentration of 95% to 99% would be used to form the chlorine salts for use as the fused salt fuel of a fast breeder reactor.

Production rates of approximately 17,000 lbs of chlorine per year can be obtained at a steady-state unit cost of approximately \$150 per lb.

Lithium Isotope Separation Facilities

Lithium-7

The Y-12 Plant has facilities designed to enrich lithium isotopes. The nuclear properties of lithium-7 together with the physical properties of lithium are such that lithium highly enriched in Li-7 is an attractive material for a reactor heat transfer fluid either as a metal or a fused salt. Li-7 also has potential application as a hydride in SNAP reactor shielding applications. Naturally occurring lithium has a thermal neutron absorption cross section of 70.9 barns. Lithium, enriched to 99.99% Li-7, has a cross section of 0.13 barns. The published unit cost is \$120 per kg.

Chemical Exchange

Boron-10

As a result of a survey on potential demands for boron-10 we prepared a preliminary design and cost estimate for a plant for the production of 5 kg. per day of boron enriched to 95 atom % B-10. The isotope separation plant utilizes a chemical exchange reaction

The unit cost of production from the isotope separation plant is \$450 per kg. of boron as BF_3 .

This preliminary cost estimate is based on the plant location at one of our production sites where such facilities as building and utility distribution systems and plant auxiliary services are available. At present, a development program including the construction of a pilot plant is in progress at ORGDP to define certain process variables, materials of construction, process chemistry, etc., more accurately. The pilot plant could be expanded into an interim production facility to produce about 2 kg. per day of B-10.

Molecular Distillation

The physical and thermal properties of potassium make its use as a reactor coolant desirable in mobile nuclear reactors. Potassium, however, when exposed to the reactor core acquires an undesirable activity of gamma arising from the nuclear reactor between neutrons and the potassium-41 isotope which is present in natural potassium to the extent of about 7%. Thus the elimination of heavy shielding and easier maintenance of the power plant can be realized from the use of such reactors of potassium from which the K-41 isotope has been substantially removed.

Cost estimates were made for the production by the molecular distillation process of 2,000 lbs. per year of potassium in which the K-41 content was reduced by a factor of 100, which corresponds to a K-39 concentration of 99.93%. The resulting unit cost was \$560 per lb. The capital investment would be \$5,000,000. The separation parameters used in this study were obtained by extrapolating the results obtained from experience at ORGDP on

lithium isotope separation by molecular distillation. At present, a development program is in progress at ORGDP to develop a prototype molecular distillation tray suitable for a plant in order to confirm the design bases.

In conclusion I would like to say that we feel that these capabilities for the separation of kilogram quantities of various isotopes at reasonable costs could lead to extensive applications in the atomic energy program. As has been indicated the modification of the nuclear characteristics of materials with desirable chemical and physical properties can make for substantial improvements in reactor designs.

THE GAS CENTRIFUGE

E. C. Evans, ORGDP

The gas centrifuge was intensively investigated during World War II for the separation of uranium isotopes. Very complex machines were built and operated individually. Because it was evident difficult mechanical development problems would have to be solved to make large scale use of the process practical, the effort was diverted to more promising methods of isotope separation.

Some research and development work was carried on in the years following, and in 1960 the effort was substantially increased. The reasons for the increased interest included the demonstration of a centrifuge having a mechanical system less complex than those developed previously and the rapid advances being made on materials which might be suitable for centrifuge rotors.

Dr. Gernot Zippe, an Austrian, came to the University of Virginia and built and operated centrifuges similar to those he developed in Russia following World War II. This centrifuge design is represented schematically in Figure 1. It consists of an aluminum rotor supported on a pivot or "needle" bearing at the bottom and a magnetic bearing at the top. Spring supports and dampers are provided for both bearings. The rotor is driven by an electric motor. The motor armature is a steel disk attached to the bottom of the rotor and the motor stator is located directly below. The rotating magnetic field created by the stator couples

* This document is based on work performed at Oak Ridge Gaseous Diffusion Plant operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

ZIPPE SUBCRITICAL CENTRIFUGE

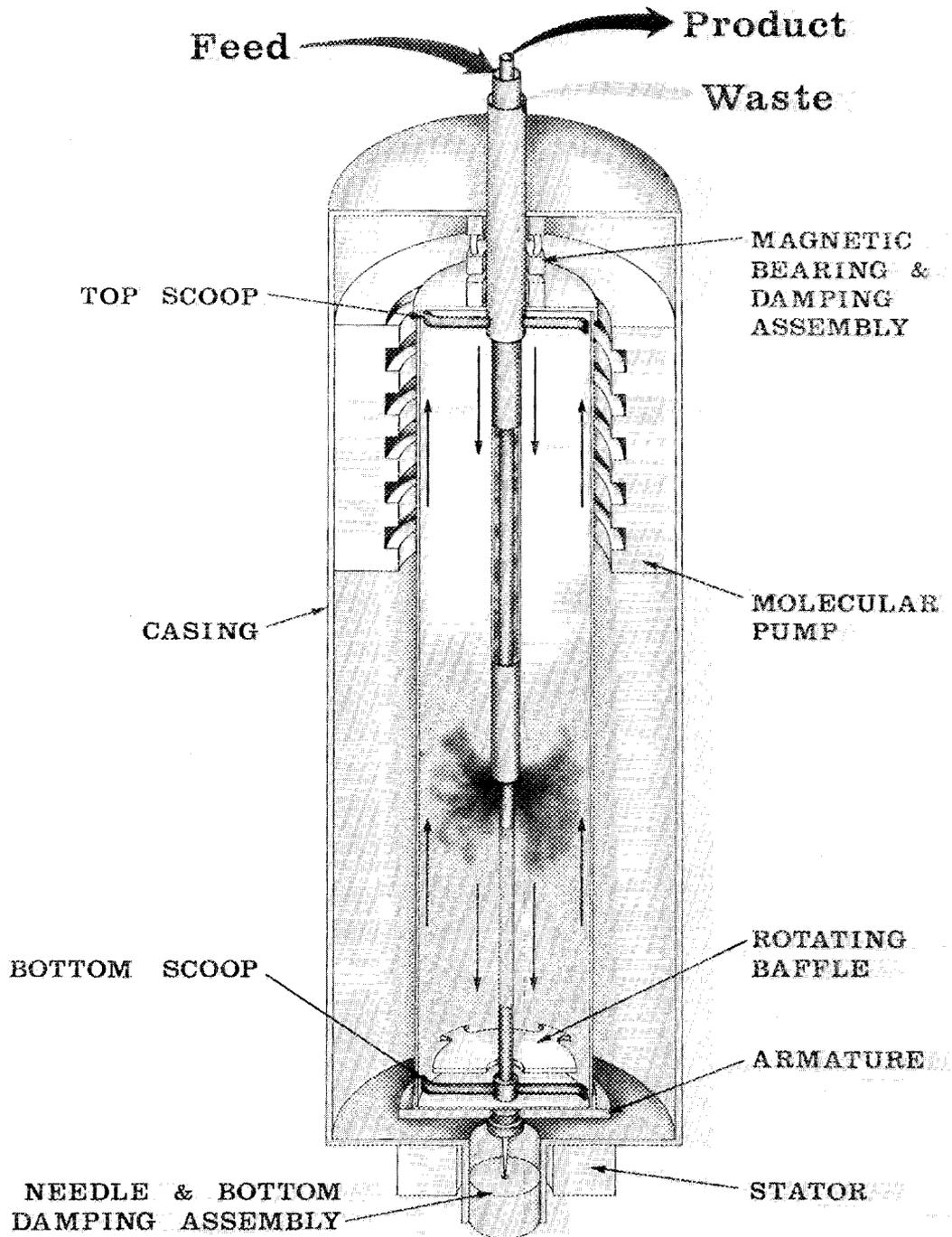


Fig. 1

to the armature and supplies the drive torque to rotate the rotor at 90,000 rpm. The centrifuge rotor operates in a vacuum to reduce heat due to gas drag. A molecular pump about the top end of the rotor pumps upward the process gas which leaks out the top of the rotor, thereby acting as a seal to maintain the vacuum on the lower part of the rotor.

Process gas is admitted, through one of three concentric tubes, to the center of the rotor. It is caused to revolve by friction with the inner wall of the rotor, and the heavier molecules are forced toward the wall by the centrifugal field and the lighter ones are displaced inward. This action can achieve one "stage" of isotope separation. By creating an internal flow in the rotor with the gas moving axially at the periphery and in the opposite axial direction at smaller radii, a cascade effect is achieved. This increases the separation factor of the centrifuge to the equivalent of several stages. The major composition gradient is axial in the centrifuge, and the gas is removed by scoops at each end. Since the centrifugal field also creates a compression ratio of the gas of about 3000, the gas is forced out the scoops with sufficient pressure to drive it to the next centrifuge in a cascade.

The Zippe type centrifuge combines the separator, pump, and seal all in one machine which is less complex than gas centrifuges developed earlier. However, it still requires precision parts and skillful assembly to obtain satisfactory operation.

The separative capacity of a gas centrifuge is, to a first approximation, proportional to the fourth power of the peripheral velocity of the rotor.

This places great emphasis on high rotational speeds for the rotors and consequently on materials having a high strength to density ratio, and which can be formed into precision rotor parts. These are the same requirements for some aircraft and missile components on which significant advances have been made in recent years, thereby providing many materials which could be of use in the development of superior gas centrifuges.

A comparison of the separation theories for gaseous diffusion and gas centrifugation is given in table I. Note that the separation depends upon the square root of the ratio of the molecular weights of the isotopes for gaseous diffusion and on the difference in molecular weights for gas centrifugation. This means that the gas centrifuge can develop a high separation factor for heavy isotopes relative to that obtained using many other processes. For example, for the $U_{235}F_6$ - $U_{238}F_6$ molecules, the theoretical ratio for one stage of gaseous diffusion is 1.00429, whereas for the Zippe centrifuge, it is 1.225 or about 50 times as large. This means that a relatively small number of centrifuges might be used to enrich natural uranium to weapons level. This is a matter of concern since there is a possibility a non-nuclear power could operate a clandestine plant for weapons production.

The attractive features of the Zippe centrifuge, advances in materials, concern over clandestine centrifuge plants, and a more complete evaluation of the potential of the process are some of the reasons for the renewed interest in the gas centrifuge process. In December, 1960, the United States Atomic Energy Commission announced that it had recently

TABLE I
 COMPARISON OF IMPORTANT PROCESS PARAMETERS FOR THE SEPARATION OF
 URANIUM ISOTOPES WITH GASEOUS DIFFUSION AND GAS CENTRIFUGATION

Process	Maximum Process Separation Factor
Gaseous Diffusion	$\alpha = \sqrt{\frac{M_2}{M_1}} = 1.00429$
Elementary Centrifuge	$\alpha = \exp. \frac{(M_2 - M_1)(V)^2}{2RT}$ <p>V = peripheral velocity of rotor R = gas constant T = absolute temperature At V = 350 m/sec. and 25°C., $\alpha = 1.076$ Or V = 500 m/sec. and 25°C., $\alpha = 1.161.$</p>
Counter-Current Gas Centrifuge	$\alpha = \exp. \frac{(M_2 - M_1)(V)^2}{2RT} \times \frac{\sqrt{2} L}{4r}$ <p>Where L = rotor length r = rotor radius $\frac{\sqrt{2} L}{4r} \cong 2.75$ for center fed subcritical machines At V = 350 m/sec. and 25°C., $\alpha = 1.225$ Or V = 500 m/sec. and 25°C., $\alpha = 1.506.$</p>

increased the United States effort on the development of the centrifuge program and that it was expected that the total effort would cost about two to three million dollars per year. Subsequent reports indicate the work has been continued at that level.

As part of the program UCC-ND was to investigate the operation of groups of centrifuges. An experimental group of centrifuges has been constructed at ORGDP. A.E.C. programmatic and security regulations do not permit further description at this time. Anyone wishing further information should make a formal request through the Production Division, A.E.C. Oak Ridge Operations Office.

Gas centrifuges of the type demonstrated by Zippe, if successfully cascaded in a moderate number, could produce a separative capacity considerably greater than that of many processes used for small scale isotope separation, and yet much smaller than that of the gaseous diffusion plants. Such a cascade could fill the existing "isotope production gap". To separate the isotopes of an element by the gas centrifuge process, a suitable gaseous compound must be used. Some requirements for the gas are given in table II.

If anyone has an isotope separation requirement which is not being satisfied (quantity, rate, or cost) by existing processes, consideration should be given to the gas centrifuge process. We shall be pleased to provide assistance.

TABLE II
CONSIDERATIONS INFLUENCING
THE CHOICE OF PROCESS GAS FOR GAS CENTRIFUGE

Vapor Pressure	>1.5 mm Hg at 25°C. Preferably <10 ⁻⁵ mm Hg at Mechanical Refrigeration Temperatures
Stability	Negligible Decomposition Below 100°C.
Corrosivity	Negligible for Aluminum, Copper, Nickel and Brass at 100°C.
Interfering Isotopes	Minimum but Subject to Discussion Dependent on Individual Separation Tasks
Molecular Weights	>150; Desired Isotope as Large a Fraction of Total as Possible
Miscellaneous Considerations	Safe Handling, Toxicity, Explosivity, Costs, Radioactivity, Nuclear Criticality

AEC COSTING AND PRICING POLICIES AND PRACTICES FOR RESEARCH MATERIALS

L. S. Lenderman, USAEC (HQ)

I would like to speak briefly about our general costing and pricing practices in AEC.

Costing

First, we will look at our costing principles and procedures. The cost of materials produced for services in connection with regular AEC research and development programs are charged as incurred to the appropriate AEC program and activity. Costs must be segregated between fund and non-fund for cost-budget reporting purposes. When materials are transferred within AEC, the costs are transferred on AEC Form 325 and are segregated between fund costs, fuel costs, and depreciation.

Now let us look at some of the categories of cost which must be considered in accounting for materials. For an example, isotope production costs shall include, as applicable, the following:

1. Direct Material Costs representing the costs of materials, including containers, that are irradiated in the production of isotopes
2. Reactor Operating Costs including the operating costs of the reactor and all its auxiliary facilities such as steam plants, pumping stations, electrical distribution systems, waste storage tanks, central service shops, etc., that are used in connection with the operation of the reactor.
3. Processing and Other Costs including the costs of chemical separation and purification of products, special shielding and handling where required.
4. Contractors' Overhead Costs including the applicable portion of the overhead that is chargeable to fuel fabrication, reactor operations, processing, etc.

All these costs require the expenditure of funds usually during the period in which the work is being accomplished. In addition, there are certain non-fund costs for which funds were previously expended such as the following:

1. Net Fuel Costs representing a proportionate share of the cost of SS materials used in fuel fabrication, transportation, fuel preparation and fuel separation and recovery, less the value of the material recovered.
2. Depreciation including depreciation expense on facilities used directly in the production and processing of isotopes plus a proportionate share of depreciation expense for contributing services. Where facilities are used only in part for the production and processing of isotopes, a proportionate share of depreciation expense for the jointly used facilities shall be included.

Isotopes produced as a by-product of research are costed at the established average costs of production for such isotopes if already a part of an inventory account. If a new isotope is produced as a by-product of research, the cost should be estimated.

Pricing

Next we shall look at our pricing policies and practices. A revised Chapter 1701 and related Appendix 1701 for pricing have just recently been issued.

Chapter 1701 contains a statement of the policy of the AEC in regard to pricing. The policy provides that materials and services furnished to others shall be priced at the higher of full cost recovery or current commercial prices so long as these prices (a) will not discourage the use of such materials and services or the development of sources of supply of such materials or services and (b) will not discourage research and development. This policy is unchanged from the preceding issuance. However, the responsibilities and authorities have been revised to some extent. The Director, Division of Industrial Participation, now consults on prices to be established and furnishes information and recommendations. Basically, the responsibilities and authorities do not change for other Heads of Divisions and Offices, Headquarters or for Managers of Field Offices.

Full cost consists of those categories of cost, direct and indirect, that I enumerated earlier plus a factor to be added which is intended to cover other measurable expenses such as process development, startup and standby, AEC administration, security clearances, etc., and unmeasurable expenses such as insurance, the AEC's share of general government administration, and facilities and services furnished by central agencies.

Specific added factors have been developed for a number of materials and services and can be obtained from the Office of the Controller, Headquarters. Where a specific added factor has not been established for a particular material or service, a factor of 15% shall be used.

Appendix 1701, Pricing, contains costing and pricing data on specific materials, a list of exceptions to the full cost recovery policy, general billing procedures, and a list of prices charged for some of the materials and services furnished by the AEC.

A recent change in one of the exceptions to our policy of full cost recovery should be mentioned. For research and development activities an agreement or arrangement for reduced charges may be made in consideration of benefits to AEC programs from such work. Previously, the determination of such an exception was made by the field office managers or the Director, Division of International Affairs, on a case by case basis. Field office managers may now approve written procedures under which such determinations of exception

may be made by the appropriate designated official of the multi-program laboratories and other laboratories. However, such activities must be capable of being performed within current budgets and manpower limitations of the involved contractor.

In closing, I would like to make a suggestion which would assist us greatly at Headquarters. In providing prices or estimates of cost for materials or services, please coordinate with your field office finance division. This will insure the consideration of all appropriate cost factors and will eliminate the need for the Controller to come back to the field for cost verification by the finance personnel.

DISTRIBUTION

H. A. Larsen, USAEC (ORO)

With all the good speakers who are on this program, there is little that they are not more able to present. It may be in order, however, to suggest a couple of points for consideration in the area of procedure.

First, it seems sometimes that the formality of an order to cover the furnishing of materials and services is an unnecessary chore which does nothing but hamper the desired activity. To this I can say that while it may be a chore, there is a need to record the terms of transfers for the protection of both sides to the transaction, and a little documentation of where costs should lie and what responsibilities are or are not accepted is from time to time quite important. The documentation of the important case is not likely to be present unless the documentation of all transactions is done routinely since, unfortunately, none of us is able to point out beforehand which transaction may later be the source of trouble.

Second, the task of documentation is not nearly so bothersome as it might be, considering that there are basically only two AEC forms for the sale of most of the research materials - Form AEC-391 for sales to private parties and Form AEC-375 for transactions between AEC cost-type contractors and Federal agencies. In the event that there are, from time to time, special cases, I am sure that each of you will be most welcome to ask for assistance from your local AEC office.

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