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REVIEW OF ISOTOPES TARGET PROGRAM

JANUARY 1965 – DECEMBER 1967

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OAK RIDGE NATIONAL LABORATORY
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UNION CARBIDE CORPORATION
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ISOTOPES DEVELOPMENT CENTER

REVIEW OF ISOTOPES TARGET PROGRAM
JANUARY 1965 — DECEMBER 1967

E. H. Kobisk

Isotopes Division

INDC-13

JULY 1969

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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REVIEW OF ISOTOPES TARGET PROGRAM
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SUMMARY

Research, development, and production activities of the Isotopes Target Center expanded greatly during this period to meet the increasing number of service requests. Technology unique to the preparation of thin film targets was broadened and was applied to the formation of ultrapure-isotope and normal-assay materials for use in nuclear interaction research and other programs more closely associated with solid state physics and metallurgy. In addition, more emphasis was placed on fabrication of targets containing the actinide elements.

Improvement in the quality of targets and in the capacity of the Center to prepare more sophisticated samples resulted in greater utilization of target preparation services. In 1964 the Center produced an average of 100 targets per month; in 1967 this average had increased to 250. Many additional samples (e.g., plates, castings, wires) which, strictly speaking, cannot be classified as targets were prepared for a variety of research programs other than nuclear physics. Roughly 25% of the total manpower of the Center was assigned to this latter function.

Support of these accelerated activities occasioned a corresponding growth, with increased depth, in the research aspects of the special materials program. More basic investigations involving electron microscopy, zone refining, single crystal growing, and metal distillations were initiated with emphasis on long-range results.

HIGH-PURITY ISOTOPE MATERIALS PROGRAM

Contaminants can cause significant changes in the physical and chemical properties of elemental isotopes used in target preparation and can reduce the accuracy and sensitivity of results in experiments utilizing the targets. The effect of contaminants can be particularly detrimental in the preparation of thin, self-supporting films. Traditional purification techniques are usually inadequate for use with enriched isotopes because of low product yields or because of the possibility of degradation of isotopic assay by contamination with normal-assay material. Therefore, methods for purifying milligram to multigram quantities of isotopically enriched material must be developed. An added benefit of the purification program is the preparation of alternative chemical forms of isotopes (particularly elemental) to those normally offered.

Reduction-Distillation of Metals

The reduction-distillation technique has been found to be applicable for the preparation and purification of many isotope metals.[1] Four critical characteristics of the metal reductant are necessary to achieve successful reduction-distillation:

(1) The reductant must react with the metal oxide at a temperature which is not too different from that at which the product metal exhibits a vapor pressure of ~1.0 torr.

(2) The reductant metal should be close to or above its melting point at the desired reaction temperature.

(3) The vapor pressure of the reductant must be negligible ($<10^{-7}$ torr) at the reaction temperature.

(4) The reductant metal oxide(s) must have a vapor pressure equal to or (preferably) less than that of the reducing agent itself.

Most metal reductants can be excluded on the basis of their vapor pressures at the temperature of reaction (depending on the metal oxide to be reduced). The most useful reductants whose properties frequently satisfy the four criteria noted above are Al, La (and other rare earths), Zr(Hf), Ta, Th, and, upon occasion, U.

The preparation of magnesium metal illustrates the dependence of the reduction-distillation method on the characteristics of the reductant. Although aluminum metal proved successful in reducing MgO, the yield of product metal from this reaction was invariably low (usually 50-60% of the theoretical amount), and aluminum contamination in the product ranged between 200 and 1000 ppm, depending upon temperature control. It was found that when lanthanum metal was used as the reductant, product yields increased to 90-95% without lanthanum contamination of the product since a higher reaction temperature (~1150°C) could be used. Lanthanum contamination of the magnesium never exceeded 50 ppm.

Most reduction-distillation reactions proceed smoothly, and the rate of product vaporization (reaction) can be easily controlled by temperature variation when the reductant metal is above its melting point. Several reductants (e.g., zirconium and tantalum powders) were found which cause reduction well below their respective melting points. Tungsten metal has this capability, but the reaction product, WO_x, is highly volatile and causes gross contamination. With each of these three "solid" reductants, there appears to be a very distinct temperature at which rapid reduction and volatilization occurs and below which no reaction is observed. The rate of volatilization is difficult to control since reduction occurs several hundred degrees above the temperature required to produce a vapor pressure of ~0.1 torr (the pressure usually employed in vacuum distillation-condensation procedures).

The reduction-distillation technique was used to prepare high-purity isotopes, in elemental form, of all alkaline earth (Group II-A) elements except radium, potassium, and all lanthanide elements except lanthanum and cerium. This process is limited to preparation of metals in quantities which cannot be obtained as efficiently or in sufficient purity by some alternative method. Table 1 summarizes the conditions and efficiencies of the reduction-distillation technique for those elements whose isotope metals are now being made available by this technique.

Table 1. Conditions for Metal Isotope Preparation by the Reduction-Distillation Technique

Metal oxide	Reductant	Reaction temp, °C	Crucible material	Heating technique[a]	Collection efficiency, %
Mg	La	1050-1100	Ta	R	87
Ca	La	1150-1200	Ta	R	85
Sr	La	1025-1075	Ta	R	85
Ba	La	1150-1250	Ta	R	58
Si	Ta		C	EB	—
Nd	Th	1950-2000	Ta	R	75
Pm	Zr	1500-1700	W	EB	—
Sm	La	1300-1350	Ta	R	78
Eu	La	1150-1200	Ta	R	92
Gd	Th	1900-1950	Ta	R	63
Dy	Th	1750-1800	Ta	R	72
Er	Th	1850-1900	Ta	R	85
Yb	La	1125-1200	Ta	R	75
Np	Zr	~1400	W	EB	—
Am	Zr	1000-1100	Ta	EB	50

[a] R — resistance (ohmic) heating; EB — electron-bombardment heating.

Elemental calcium isotopes were initially prepared for accelerator and solid state physics research by reduction of CaO with lanthanum metal and subsequent (or simultaneous) distillation of the product metal from the cermet compact. When large quantities (1 to 100 g) of metal were required, a modification of the process was found applicable — "fractionation" of metal vapors. By using a distillation column integrated with and above the reaction zone, as illustrated in Fig. 1, higher reaction temperatures could be employed with associated increased yield of product metal. The perforated plates in the column were maintained at a temperature lower than that of the reaction zone so impurity metals with a relative vapor pressure lower than calcium were condensed, and increased purity of the product was achieved. The requisite temperature gradient up the column was created by varying the number and spacing of the helical induction coils surrounding the reaction-distillation vessel. Figure 2 illustrates this technique as applied to the preparation of 65 g of ^{40}Ca metal. A total impurity content of <300 ppm by weight was achieved with a product yield of 95%. Eventually, this product metal was pressed into 1-in.-dia pellets for neutron scattering experiments.

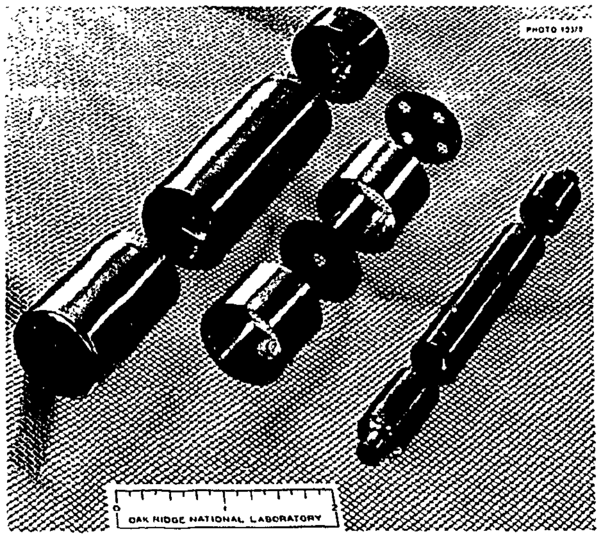

Fig. 1. Metal Reduction-Distillation Column. (A) Top section is effusion port, center section is distillation column containing perforated plates, and lower section is reaction vessel. (B) Disassembly of distillation column components. (C) Similar still used for <0.5-g quantities of material.

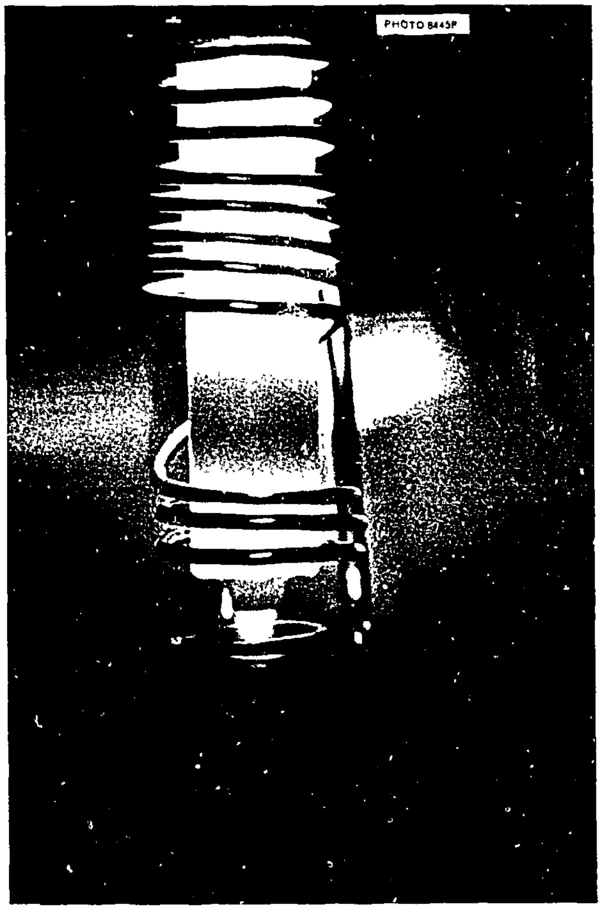

Fig. 2. Reduction-Distillation of Isotopically Separated Calcium-40 in an Induction-Heated Tantalum Column. Differential heating of the column allows purification by fractional distillation. Ionized calcium metal vapors are issuing from the top of the column.

The reduction-distillation method can also be used to separate a specific element from a mixture of other elements. A combination of Eu, Gd, and Sm in the ratio 100:20:5, respectively, was fractionated in a tantalum column. After a single reduction-fractionation, the purity of the europium was >99.5% The gadolinium and samarium components were retained in the reaction mixture. Similarly, the purity of some actinides can be enhanced by application of this technique, particularly where fission products are in combination with the desired element.

Application of the fractionation technique in the preparation of ultrahigh purity ^{151}Eu and ^{153}Eu metals permitted the fabrication of elliptical targets weighing 10 g each and having major and minor axis dimensions of 3.25 and 2.25 in., respectively. These targets were cold-rolled in a purified argon atmosphere to inhibit oxidation.

Other high-purity lanthanide metals were prepared by the reduction-fractionation technique. Cold-rolling of various isotope (or normal) metals derived in this manner resulted in foils with a minimum areal density of ~800 μg/cm^2 (0.0008-0.0012 in.) for an area >6 cm^2 and ~500 μg/cm^2 for ~2-cm^2 area. The oxygen and nitrogen content of these metals was usually <500 ppm.

van Arkel-de Boer Purification of Isotopes

The van Arkel-de Boer purification process[2] has been used commercially to obtain high-purity Group IV-B metals. Usually poor yields (~20-50%) result in the manufacturing-scale process; however, a modified process has been developed which permits purification of small quantities of these metals without loss and with product yields of ~75-95%. For purification of isotopic material, a Pyrex glass reaction vessel of ~1-liter capacity is used (see Fig. 3). The purified metal is collected on a 0.007-in.-dia tungsten filament attached to the two tungsten electrodes sealed into the top of the vessel.

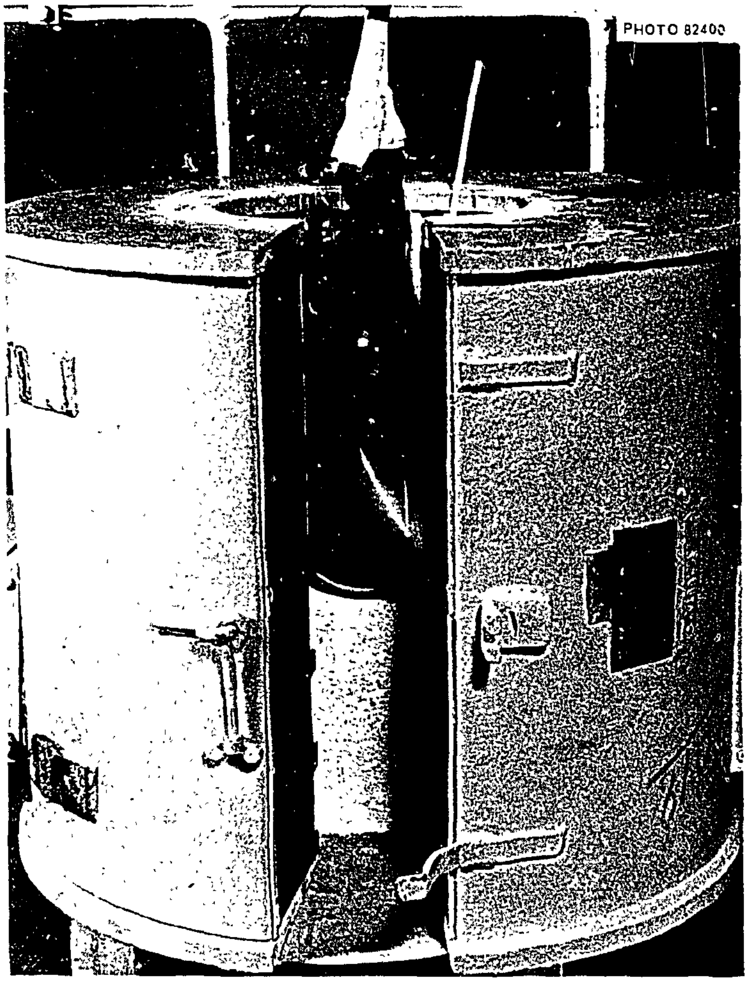

Fig. 3. A van Arkel-de Boer Reaction Vessel and Furnace for Purification of Zirconium, Titanium, and Hafnium Isotopes.

In operation, the impure metal (usually as powder) is poured into the vessel to a level which is out of contact with the tungsten electrodes. Since powdered samples have high surface area and considerable sorbed gas content, long outgassing periods are required to desorb and remove the bulk of these impurities. If not removed, these gaseous impurities can cause contamination and loss of metal ductility by being sorbed in the crystal bar during the process. Usually a vacuum level of 2×10^{-9} torr is achieved in the reaction vessel before it is sealed.

Addition of purified iodine from a sealed glass ampule broken inside the reaction vessel initiates the purification process. The entire sealed vessel is placed in a furnace and maintained at the appropriate iodine-metal reaction temperature. The tungsten filament is heated to the requisite temperature, and thermal deposition proceeds until no further growth is evident. Figures 4 and 5 illustrate the growth process and the resultant bars of material obtained by this technique.

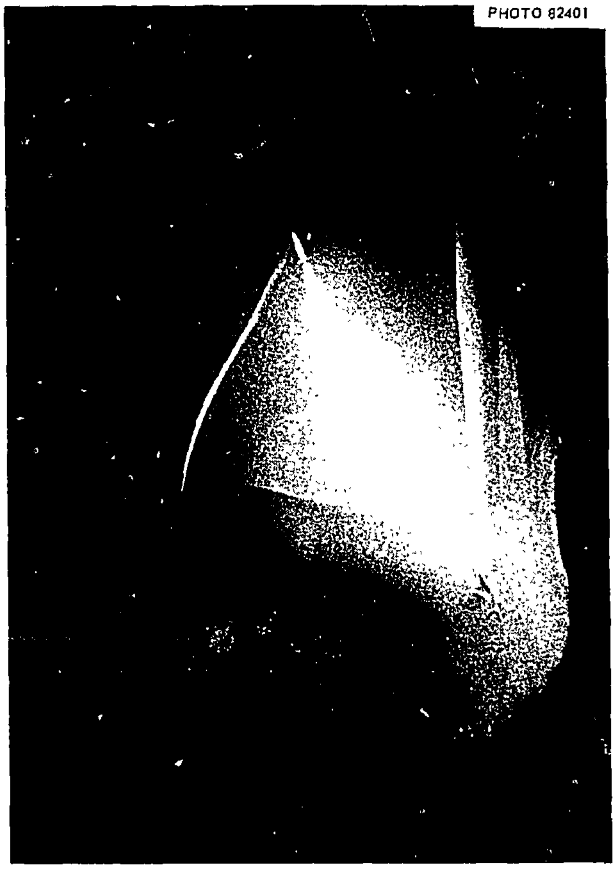

Fig. 4. Hot Filament Portion of van Arkel-de Boer Reactor. Crude titanium metal sponge and iodine crystals can be seen at the base of the reactor.

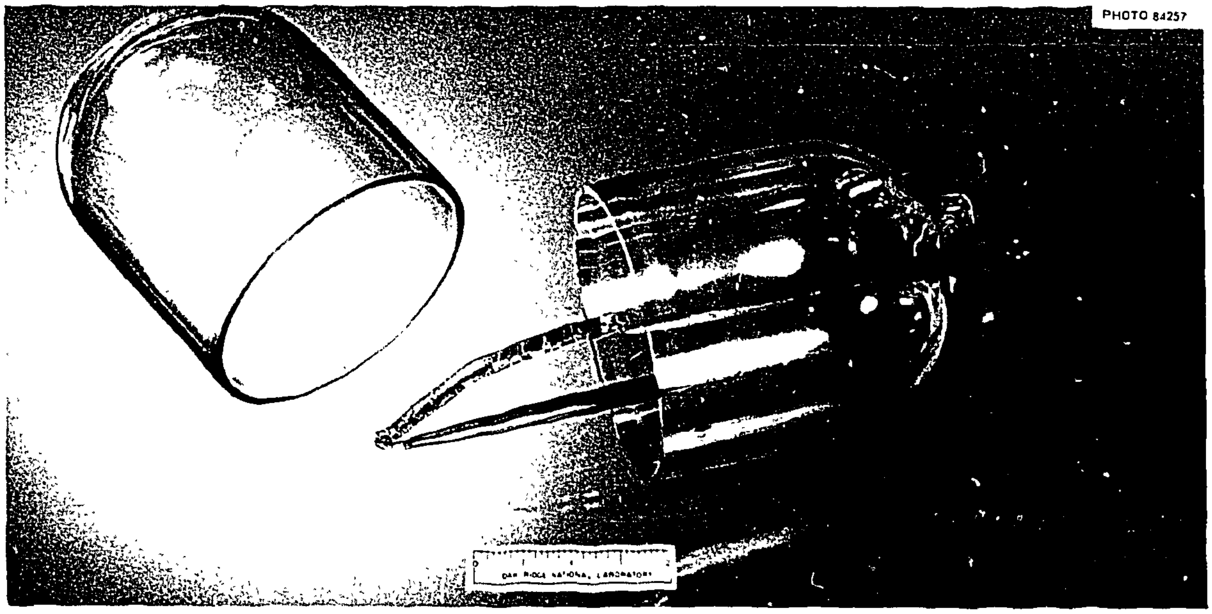

Fig. 5. Crystal-Bar Zirconium Metal as Formed on the Tungsten Filament.

When crystal bar is employed as starting material for mechanical rolling of thin foils, the metal is cracked from the tungsten filament and the remaining pieces of tungsten are removed under a microscope. Unfortunately, the layer of metal surrounding the filament is usually highly contaminated with tungsten. The depth of this layer is unknown, and the tungsten is generally not removable from the bulk of the material. When titanium is being prepared, the tungsten filament is completely dissolved by the resulting crystal bar.

Several techniques were developed to eliminate tungsten contamination. The two techniques found most applicable were (1) use of increased filament temperature to form an annular void around the filament and thus prevent tungsten diffusion and (2) use of a filament of the same isotope and composition as that of the crude metal to be purified, thus eliminating all tungsten from the system.

By increasing the tungsten filament temperature about 200°C above that necessary to rapidly decompose the metal iodide, thermal cracking apparently occurs prematurely in the vicinity of the filament rather than directly on its surface (Fig. 6) resulting in the requisite annular deposit. This procedure was found effective for producing material that could be easily separated from the tungsten in small pieces.

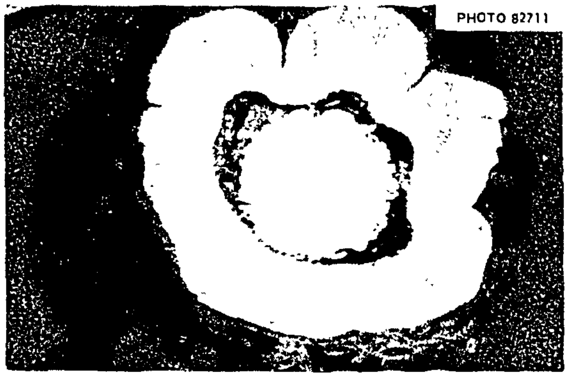

Fig. 6. Annular Deposit of Zirconium on Tungsten Showing Superficial Connection Between Deposited Metal and the Filament.

From purified material obtained by the annular deposition technique, it is usually possible to obtain a single piece of metal which can be extruded into wire a few inches in length. This wire can then be substituted for the tungsten filament, and a crystal bar grown using the wire as the nucleus. Resultant bars of metal are then homogenized and further purified by zone refining under high vacuum. Once this latter bar is obtained, similar filaments, devoid of tungsten, can be fabricated easily, and additional bars can be obtained by repeating the procedure.

Bars of zirconium isotopes 90, 92, and 94 were prepared in this manner for study of the electrical inversion temperature dependence upon isotopic content. In this case it was found that a 24-hour anneal (at 800°C) of the zone-melted bars was required to obtain a sharp transition from normal to superconductivity. The metal bars (1.5-in. length; 0.2-in. dia) were found to contain no more than three macrocrystals (as twins) in each bar. Similar bars of titanium even-mass isotopes are now being prepared.

The van Arkel-de Boer process has been used to purify Ti, Zr, Hf, Cr, and Nb metals and their associated isotopes. Purities of >99.9% are almost always attainable with the major impurity component being tungsten (from the filament). When crystal bars are grown on filaments prepared from the same isotope, purities of 99.99+% are easily obtained. Most metals exhibit enhanced ductility after processing in this manner and have been cold-rolled into foils with areal density of <500 $\mu g/cm^2$; chromium is the single exception. Crystal bars of several metals are shown in Fig. 7.

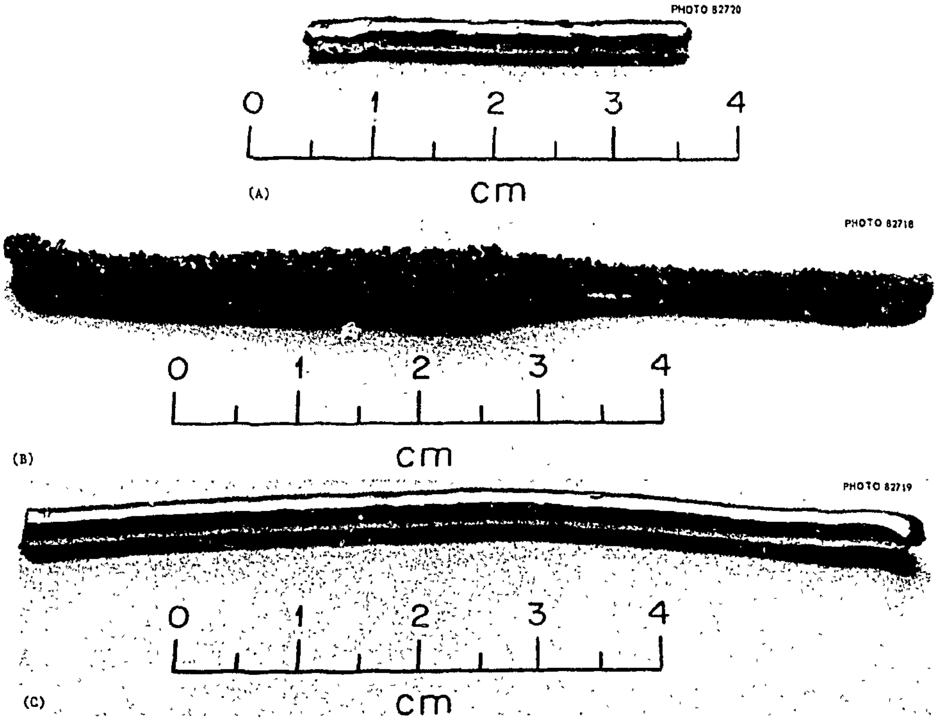

Fig. 7. Crystal Bars of (A) Zirconium, (B) Niobium, and (C) Titanium. Note large crystal faces in each bar; these may be used as seeds for single-crystal growing.

VACUUM EVAPORATION TECHNOLOGY

Vacuum evaporation is a versatile technique for preparing self-supporting films of elements in almost any thickness range up to 50,000 A and supported films on almost any substrate material. Each element and film thickness poses preparative problems unique to the material and is treated as an individual development problem in vacuum evaporation technology. Techniques suitable for measurement of film thickness or uniformity of film thickness must be included as an integral part of the development effort. Since this is a summary report, details on each film preparation are not included, but the results of these developments will be discussed. For general information, a listing of routinely prepared samples is given in Appendix A.

Self-Supported Films

In the past few years, improvements in vacuum evaporation techniques and improved vapor condensation methods have permitted attainment of a wider range of film thicknesses and sizes. Generally, films of more reactive metals (e.g., rare earths) have been prepared in association with the reduction-distillation process described earlier.

Bismuth

Preparation of accelerator targets from elemental bismuth to achieve large self-supporting areas of >13 cm^2 and high areal densities was successful only when vacuum evaporation techniques were used. Since bismuth is monoisotopic and relatively inexpensive, evaporation-condensation could be carried out at large vapor source-to-substrate distances to achieve high uniformity. By using a detergent as the parting agent, condensed films of 30 cm^2 were prepared in self-supporting form having areal densities between 3-12 mg/cm^2 simply by evaporating the element from a tantalum boat using resistance heating. At these areal densities the films will not float on water since their weight exceeds surface tension. If, however, the substrate and film were soaked in water, the physical strength of the bismuth was sufficient to withstand the tension required to slip the film from the substrate onto a mounting frame of suitable dimensions. Thinner films having areal densities of <1 mg/cm^2 could be stripped from their substrates by flotation on water. A uniformity of ±5% was attainable without difficulty, and few pinholes were observed.

Germanium

For more than two years, considerable effort has been devoted to the development of a reproducible method for preparing self-supported films of elemental germanium having areal densities between 50 and 1000 μg/cm^2. Problems encountered in this preparation were associated with attainment of sufficient film ductility for nondestructive handling (particularly when areal densities >500 μg/cm^2 were required) and with elimination of pinholes in the films. When germanium vapors were condensed onto a variety of substrates, using a vapor-deposited layer of NaCl as the soluble parting agent, resultant films were found to crack radially and generally fragmented on being stripped from the substrate. Substrates of many metals and glasses were used with a variety of parting agents (ranging from detergents to KCl) without significant success. The most satisfactory procedure was to use semipolished tungsten sheet. By taking precautions to cool the substrate and to minimize radiant heat transfer between the vapor source and the substrate by use of multiple heat shields, films of good ductility were obtained. Vapor-condensed NaCl was used as the parting agent. By using 250 mg of germanium metal, reproducible films having areal densities between 950 and 1000 $\mu g/cm^2$ were produced over an area of 40 cm^2. The degree of ductility and structural strength at this thickness permitted mounting of self-supported films over a 1-in.-dia area.

Manganese

Achievement of high areal density foils of manganese (>2 mg/cm^2) in self-supporting form required considerable development. Manganese in the alpha form (<700°C) is extremely hard and brittle, and it was easily fractured during mounting. This problem was circumvented by forming a vapor-condensed layer on chilled tungsten sheet (polished) coated with a vapor-deposited layer of NaCl (~1-2 $\mu g/cm^2$). By this technique, foils with areal density up to 9 mg/cm^2 (the greatest areal density attempted) were stripped without fracture by the technique described for bismuth; the maximum area of samples produced was 6 cm^2.

Silicon

Efforts to prepare silicon films in the areal density range of 100-2000 $\mu g/cm^2$ by the reduction-distillation method failed because of the high oxygen concentration remaining in the silicon films. Tantalum metal had been used as the reductant, and the high volatility of the SiO species probably resulted in the observed oxygen contamination. However, evaporation of silicon from graphite tubular crucibles using electron-bombardment heating[1] did result in successful preparations. Compatibility of the silicon with the graphite was poor, but rapid evaporation caused only insignificant loss of isotopes by carbide formation. Silicon vapors were condensed on salt-coated tungsten sheet heated to >350°C. This minimum temperature was required to obtain suitable epitaxy resulting in films resistant to fracture during stripping and mounting. Poor results were obtained on cold substrates (amorphous films) because of significant flectional weakness of films having an areal density of >100 $\mu g/cm^2$.

Supported Films

Highly adherent coatings of isotopes or normal materials on a variety of substrates were developed for special projects during the past few years. Only a few of these which required development of special technology will be discussed.

Preparation of Magnetically Oriented Iron Films

The preparation of iron films having specified thickness and magnetic orientation was studied in order to produce specimens suitable for experimental investigation of spin polarization of electrons at the Fermi surface in ferromagnetic materials. Measurement of tunneling resistance between two similar ferromagnetic films was to be performed when the films were magnetized in parallel and antiparallel orientation; the value of the ratio of these resistances (other than unity) would be indicative of partial polarization of electrons at the Fermi surface. Samples were desired having a sandwich structure consisting of a film of iron, a layer of MgF_2 or SiO, a second layer of iron, and, finally, a cover layer of the dielectric over the entire surface.

These samples were fabricated by evaporation-condensation in a vacuum of $\sim 10^{-8}$ torr using only high-purity (>99.9%) materials. To ensure mobility of the condensing atoms (molecules) of iron or of the dielectric, the Pyrex glass substrate was heated to 300°C during deposition. All requisite evaporations were performed during a single closure of the vacuum chamber so that exposure of intermediate films to air might be avoided until the final overcoating of dielectric material was complete. To achieve magnetic orientation of the iron films, fixed Alnico magnets were placed on each side of the glass substrate to generate a field of ~20 gauss parallel to the plane of the substrate during deposition. Having the substrate temperature at 300°C enhanced adhesion of the films to the substrate and provided appropriate annealing conditions for the iron deposits.

Figure 8 illustrates the ion-pumped evaporator system used to perform these various operations. The system had a pumping speed in the evaporation chamber of ~20,000 liters/sec and was completely bakeable at 450°C. This system maintained very low hydrocarbon contamination in the evaporation chamber compared with other systems utilizing oil-diffusion pumps and cryotrapping. As shown in Fig. 9, electron-bombardment heating devices were employed to evaporate all components of the multilayer sample. Iron was evaporated from a pressed pellet of high-purity powder by driving electrons from a ring filament, immediately above the pellet, into the evaporant; this arrangement achieved volatilization in nearly 2π geometry. By locating the substrate 8 in. above this wide-angle vapor source, excellent uniformity of deposition was achieved over the desired 2-cm-dia spot.

Multiple masking of the substrate was necessary so that each iron deposit overlapped only one of the gold electrical contacts (formed by vapor condensation in vacuum) and the dielectric deposits completely covered each iron layer. The mounting jig and masking apparatus are shown in Fig. 10. A rotary mask, having three openings, could be indexed over the substrate by manipulation of an ultrahigh vacuum, rotary feed-through located on the top of the vacuum chamber. The substrate was rigidly mounted on a surface heated by a tungsten filament (Fig. 11). Near the bottom of the assembly, as shown in the figure, a quartz crystal resonator was attached adjacent to the substrate so that the mass of material being deposited during any evaporation could be monitored accurately by measuring the frequency shift of the resonator with reference to a fixed-frequency oscillator. Because

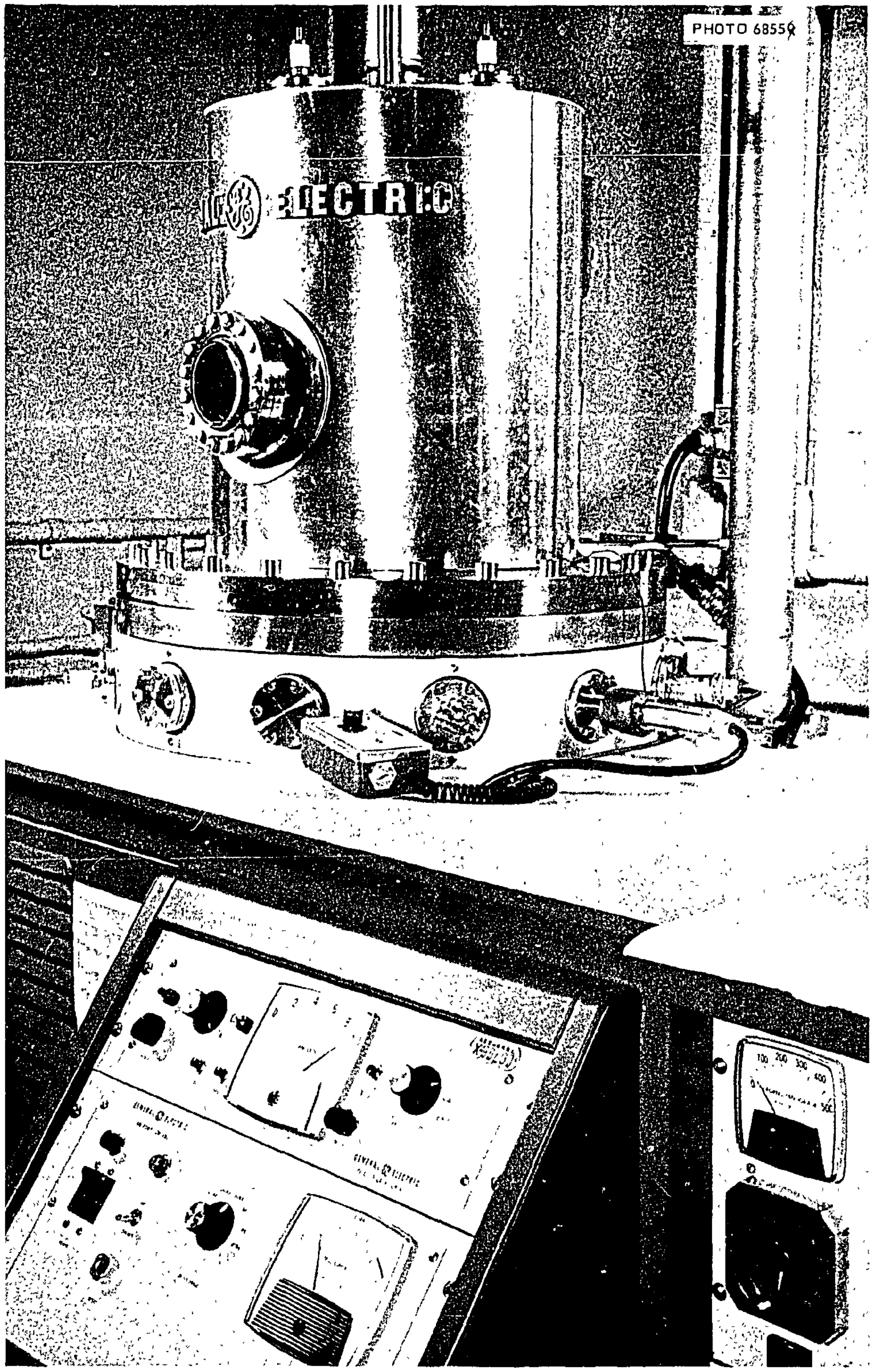

Fig. 8. Ultrahigh Bakeable Vacuum System Using Combined Titanium-Sublimation and Ion Pumping. Pumping speed is 20,000 liters/sec in the vacuum chamber; ultimate vacuum ~10^{-11} torr.

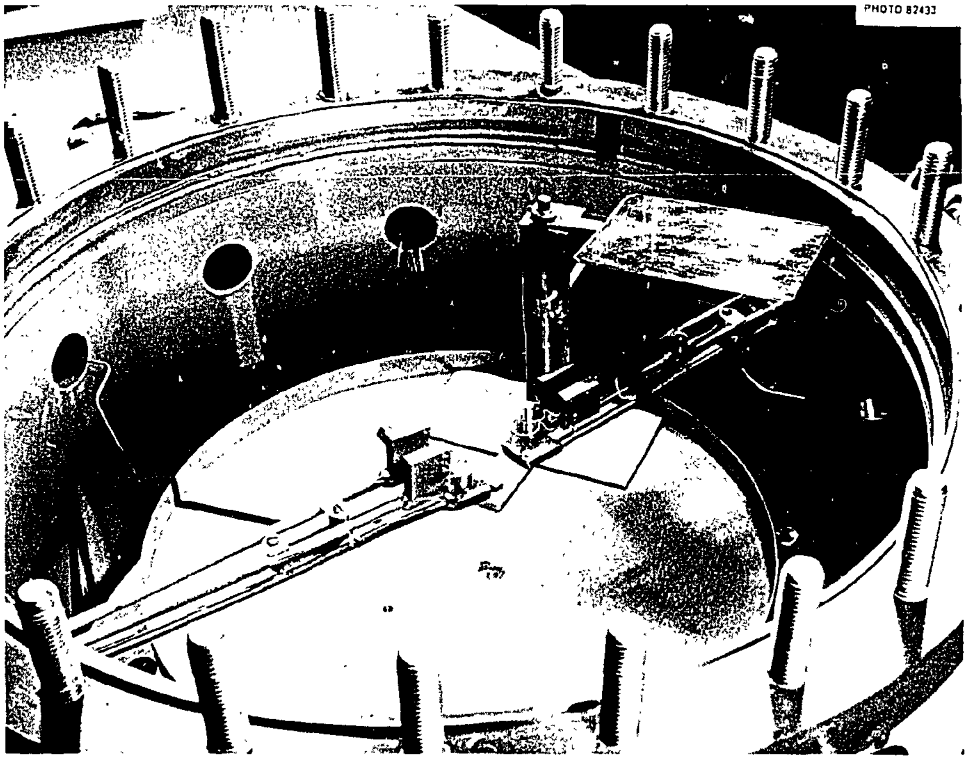

Fig. 9. Electron-Bombardment Evaporation Sources. Left-hand source uses direct electron bombardment of an iron pellet which then serves as its own crucible. Right-hand source uses a tubular crucible and effusion port for evaporation of the dielectric material.

of the wide evaporation angle and the distance between the source and the substrate, no significant difference, as determined by weight measurement, was detected between the thickness of deposit as measured by the quartz resonator and that on the glass substrate. The two Alnico magnets can be seen bracketing the substrate in Figs. 10 and 11. Since the entire assembly was constructed using nonmagnetic stainless steel, the magnetic field was parallel to the substrate and had maximum strength.

Thickness measurements using the resonating quartz crystals were found to have some temperature dependence. After suitable calibration, however, the estimated error of measurement of mass was found to be $\sim 3\%$. Temperature variations were minimized by housing the resonating crystal in a metal shell so only a spot of small, but known, area was exposed to the vapor beam. Even with this precaution, part of the 3% measurement error could be attributed to temperature variation. A finished sample of the sandwiched films is shown in Fig. 12; the iron layer, deposited directly on the glass substrate, had a thickness of 800 A, the separating layer of SiO about 100 A, and the second iron layer 1600 A. The entire sample was overcoated with >200 A of SiO. Eight similar samples have been prepared in a variety of layer thicknesses.

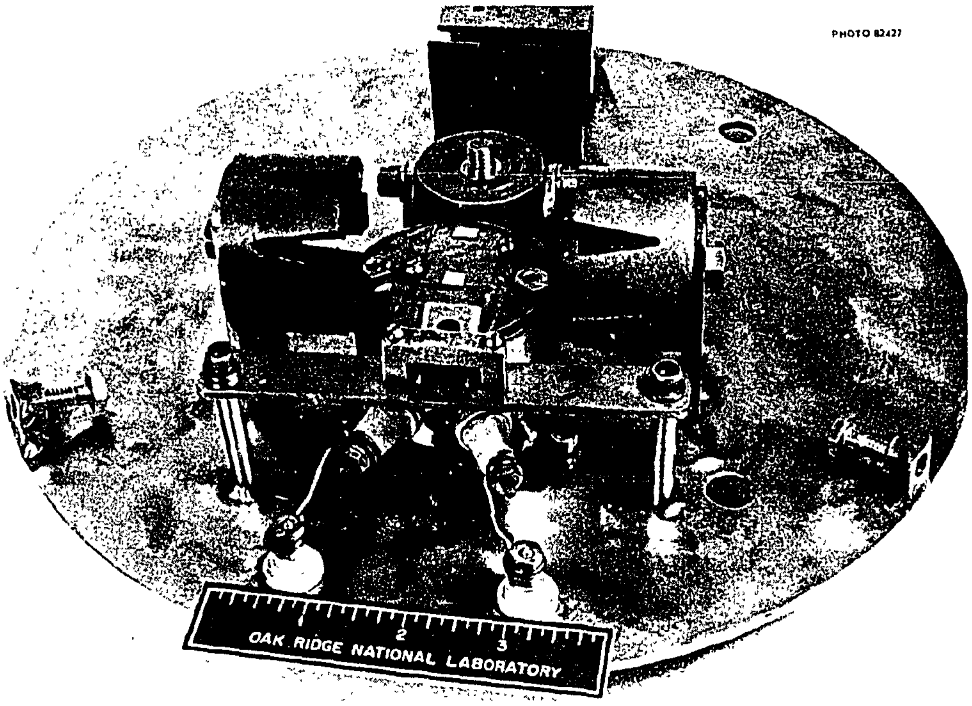

Fig. 10. Fixture for Masking a Quartz Substrate onto Which Alternate Layers of Iron and Dielectric were Deposited in a Magnetic Field. Indexing mask is shown as the rotary plate.

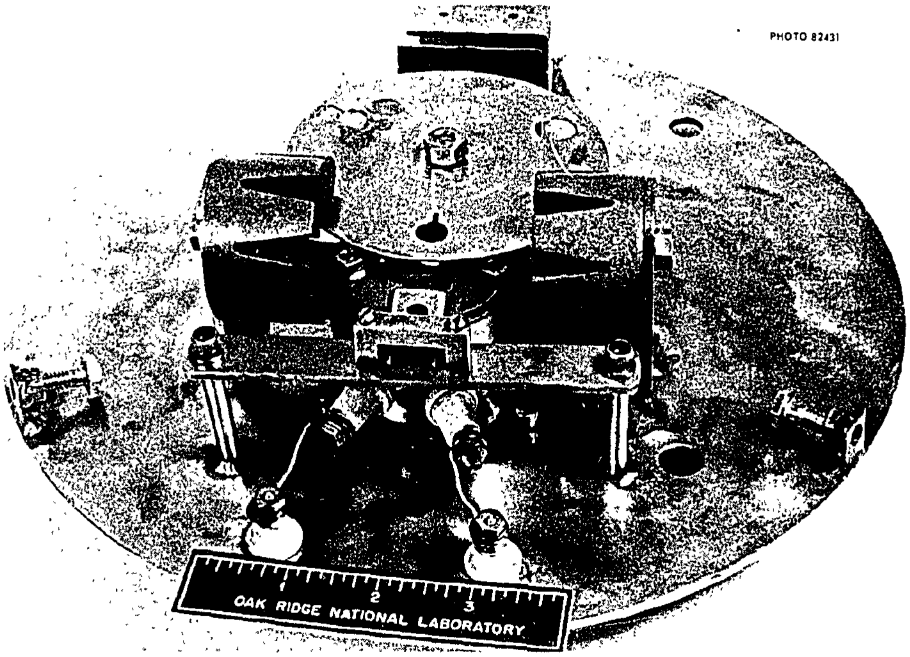

Fig. 11. Substrate Heater with Substrate in Place and Quartz Crystal Resonator for Measuring Film Thickness. Indexing mask has been removed.

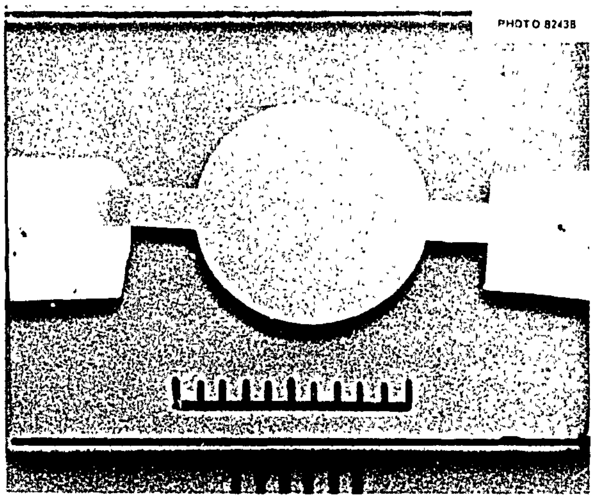

Fig. 12. Magnetically Oriented Deposits of Iron Separated by Dielectric Films on a Quartz Substrate. This sample was used for tunneling experiments in solid state physics.

Preparation of Thin-Film Platinum Resistance Telethermometry Devices

Measurement of thermal transients created on aerodynamic surfaces by movement of shock waves across the form requires the use of precision fast-response devices whose contour and mounting do not affect the surface smoothness of the device being tested. Telemetry devices such as thermocouples or thermistors have thermal response times which are too long for this purpose.

To produce a millisecond response device, 0.250-in.-dia quartz disks were employed as substrates onto which a vapor-condensed strip of high-purity platinum was formed (resistance of 100 ± 20 ohms). Provided that the film was continuous and uniform, nearly linear response to thermal transients could be observed because of the resistivity dependence of platinum on temperature. However, pinholes in the film or nonuniformity of the platinum strip could cause failure of the device and nonlinearity of response. Impurities in the film were found to be equally detrimental.

Assurance of high-purity platinum deposits was made by electron-bombardment heating of platinum metal mounted on a tungsten wire so the platinum created its own crucible. A heated substrate holder was machined to accommodate 12 quartz disks. A slotted mask through which the platinum vapors passed could be indexed over the holder. Use of 30°-tapered edges on each slot in the mask permitted deposits to be formed with nearly uniform thickness to the edge of the strip. A substrate-to-vapor source distance of 5 in. was needed to attain >95% deposit uniformity. Pinholes in the films were avoided by premelting the platinum in high vacuum and by establishing a uniform evaporation rate of the metal before opening a shutter masking the substrate holder. Maximum adhesion of the platinum to the quartz was attained at substrate temperatures of >400°C.

After platinum deposition, each disk was overcoated with a dielectric film having an areal density of ~100 μg/cm^2. Experimentation with MgF_2, SiO, and SiO_2 proved SiO to be the most satisfactory material for this purpose. Before dielectric deposition, silver metal electrical contacts were painted onto the sides of each disk and were extended to the back face for later welding of 0.002-in.-dia wire connectors. Each disk was heated in air to 400°C to cure these painted surfaces. As the final step, the SiO film was deposited over the entire surface. Figure 13 illustrates several of the finished telethermometry devices.

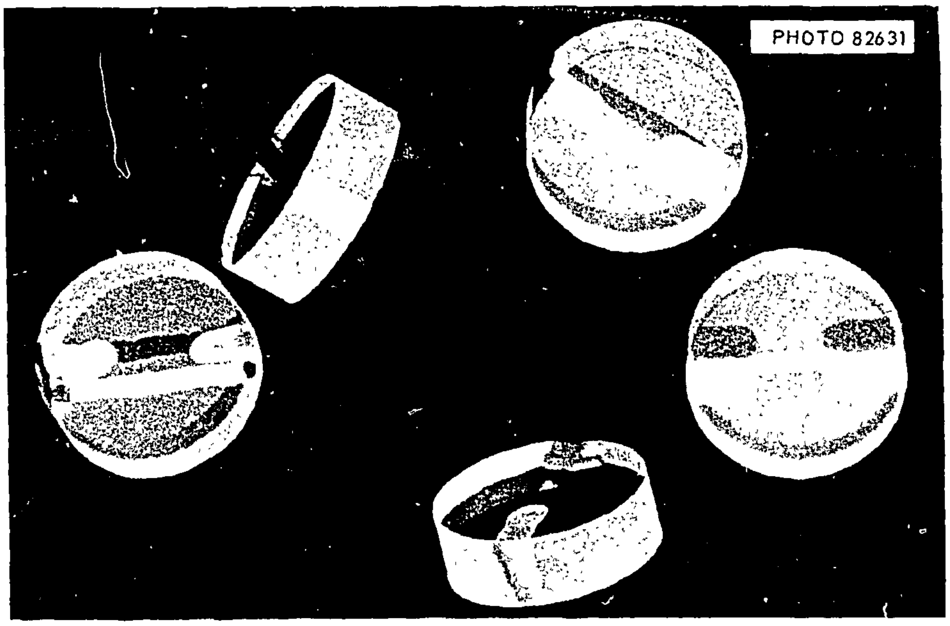

Fig. 13. Telethermometry Devices Formed by Vacuum Evaporation-Condensation of Platinum onto 1/8-in.-dia Quartz Substrates.

Tellurium Coatings on Stainless Steel and Other Substrate Materials

Tellurium is considered a nonmetal in most of its characteristics and has a high vapor pressure (10^{-1} torr at 433°C). Since the adherence of metal coatings on surfaces depends largely on the amount of diffusion bonding or chemical interaction between the coating material and the substrate, vapor-deposited layers usually are most adherent if formed at substrate temperatures in excess of 350°C. With tellurium, such substrate temperatures would be prohibitive since the "sticking" coefficient of tellurium would be much less than unity because of its high vapor pressure. Thus, development of techniques applicable to the formation of adherent tellurium coatings was necessary.

Diffusion of tellurium into stainless steel or glass at low temperatures (<100°C) was found to be too slow for good adherence of vacuum evaporated-condensed films; coatings prepared at these temperatures were easily removed by gentle wiping (no abrasion). Formation of a thin layer of an active metal telluride seemed the most feasible approach to the solution of this problem provided that a suitable active metal could be made to adhere to the desired substrate and that the quantity of this metal (thickness) was insignificant for whatever purpose the finished sample was to be used. Only those metals that could be vapor-deposited at sufficiently high substrate temperatures to form adherent chemical or diffusion bonds with the substrate were considered. An obvious choice of such metal would be one of the rare earths since rare earths easily form tellurides and tend to form highly adherent coatings on most clean metal surfaces.

Deposits of erbium metal, as little as 50 $\mu g/cm^2$, on stainless steel or glass were found sufficient to permit formation of adherent deposits of tellurium up to 2 mg/cm^2 areal density. As might be anticipated, however, this multilayered coating was similar to a graded glass-to-metal seal, i.e., the gross properties of the tellurium deposit were those of the element and not of the minute amount of erbium telluride formed at the interface. Thus, the tellurium coatings were easily removed (for all practical purposes) in vacuum by heating the substrate to >200°C. For samples to be employed at temperatures <200°C, the tellurium deposits were found to be adherent, hard, and durable, and the abrasion resistance proved to be similar to that of the bulk material.

Application of this technique in the preparation of elemental or oxide deposits of radioisotopes (e.g., the actinides) on incompatible substrates has been found entirely successful. Of particular importance has been the use of flash-coatings to form adherent bonds between hydrides and metal substrates having widely differing expansion coefficients or crystal lattice parameters. (See discussion of the HENRE tritium target preparation.) In general, the use of chemical bonding to form adherent coatings of nonmetals on almost any surface has general applicability.

Rolling of Metals

As a general preparative method for isotope targets, mechanical rolling of high-purity metals is the preferred technique whenever feasible. Rolled foils have identical physical properties to those exhibited by bulk material with the exception of microscopic crystallite distortion and work-induced strains. Both of these physical deviations can usually be removed by appropriate annealing of the foil. The material strength and uniformity of thickness (when compared with evaporated-condensed films) which can be obtained by rolling small quantities of metals are advantageous. Since little or no loss of material is experienced in forming films by cold-rolling, this technique is usually preferred for isotope processing.

Over the past three years, numerous improvements in rolling facilities and techniques have permitted preparation of foils sufficiently thin, in some cases, to be transparent. These developments have not been restricted to stable isotope target preparation but have been used with marked success in forming thin foils of many of the heavy elements, particularly uranium and plutonium.

Stable Isotope Foils

With the advent of high-purity metal preparation by the simultaneous reduction-distillation technique, many metals have been made available with the required qualities for cold-rolling into thin foils. Application of the van Arkel-de Boer iodide process for obtaining metallic titanium and zirconium isotopes, free of tungsten, also has permitted fabrication of larger and thicker samples of these metals by cold-rolling.

The use of metals produced by the reduction-distillation process (lanthanum or thorium reduced), which has improved hardness and ductility compared with commerical materials, permitted cold-working the rare earth metals into thin foils. All rare earth metals (excluding cerium and lanthanum) were cold-rolled into foils of $>6\text{-cm}^2$ area with minimum areal density of ~ 0.8 mg/cm^2 (~ 0.00003 in. thickness), and most could be reduced to areal densities of 0.3-0.5 mg/cm^2 provided the sample size was restricted to ~ 1 cm^2. Chemical reactivity of these metals toward oxygen and nitrogen is greater than for bulk materials and all rolling to thickness of <0.005 in. was performed in a purified argon atmosphere containing no more than 5 ppm of these gases.

Similarly the more reactive Group II-A metals were prepared in thin-foil form. A significant example of this preparative technique was rolling of barium isotope metal (reduction-distillation produced) into self-supporting foils of $\sim 2\text{-cm}^2$ area and having an areal density of 0.5 mg/cm^2. These foils were sufficiently thin to be transparent to strong light. Magnesium metal, being somewhat harder than heavier elements in the same group, was rolled into foils having minimum areal density of 0.1 mg/cm^2 (0.00002-in. thickness).

The minimum areal densities attainable for these various metals by cold-rolling techniques are approaching (and in some cases are well within range of) the thicknesses required for target foils used in charged-particle interaction studies. As may be seen in Appendix A, many metallic elements can be produced in self-supporting form in this thickness region by cold-rolling. The gap which existed only a few years ago between the maximum thickness of self-supporting targets obtainable by vacuum evaporation-condensation techniques and the minimum thickness to which the same metal could be cold-rolled has, in many cases, disappeared. Future development efforts will be directed toward the design, construction, and application of a miniature "cluster" mill which has small-diameter work rolls and is compatible with vacuum-inert gas environments. With such a mill, it is expected that larger area foils will be prepared in the areal density range of 0.1-0.5 mg/cm^2 for most pure metals.

Radioisotope Rolled Foils

As with stable isotope metals, the physical properties of radioisotopic metals are related to the purity of the metal. Inclusion of oxides, nitrides, or fluorides (and some metallic contaminants) causes embrittlement which precludes preparation of thin self-supported foils by cold-rolling. Because of limited processing facilities, especially shielded enclosures, all work in radioisotope metal foil preparation has been confined to alpha-emitting isotopes which can be obtained in high chemical purity; of these, only uranium and plutonium isotopes will be discussed.

In cooperation with the Center, the Development Division (Y-12 Plant, Oak Ridge) and Rocky Flats (Dow Chemical Company) personnel developed chemical conversion and reduction techniques for uranium and plutonium isotopes, respectively, that permit as little as 5 g of oxide to be processed into metallic form suitable for cold-rolling. The process converts UO_2 and PuO_2 to the fluoride, and a subsequent bomb reduction of the fluoride, using calcium and iodine, produces consolidated metal. Although this process is quite similar to that used in the production of large quantities of these metals, many developments were necessary to permit 5 g of material to be produced with 75-90% yield and attainment of a high-purity product compatible with preparation of thin foils. The details of the plutonium process can be found in a published report by Conner[3]; the work on uranium is not yet published.

To achieve conditions similar to those described for rolling stable isotope metals, an 8-in., 4-high Stanat rolling mill with dc drive was enclosed in a containment glove box. The atmosphere inside the box was purified argon continuously circulated over hot copper turnings, hot titanium-zirconium alloy turnings, and through a molecular sieve, moisture removal tray. Entrance and egress to this enclosure was only through plastic bagging ports to avoid direct exposure to the external atmosphere. A second rolling mill of the 2-in., 2-high motorized type was enclosed in a vacuum-inert atmosphere glove box.

Over the past year many samples of uranium and plutonium isotopes were rolled into thin foils. Until this facility existed, the thinnest uranium foil rolled, to the author's knowledge, was ~0.0005 in. and plutonium had not been rolled to below 0.001 in. The lower thickness limit attained by Center personnel on both of these metals is 0.00004 in. or an areal density of ~2 mg/cm^2. Because of the high density of these metals, the foils are not transparent to light at this thickness level. The reactivity of both elements is extreme at thicknesses below 0.002 in. Spontaneous ignition can occur on exposure to the atmosphere, and all rolled samples are packaged in sealed glass ampules under vacuum or in a purified argon atmosphere.

As with other thin foils, the maximum area of a rolled foil is dependent on foil thickness. However, both uranium and plutonium isotopes (when pure) have excellent flectional and ductile properties, and foils of >6 cm^2 have been prepared even at the low thickness limit. Several circular foils of 99+% ^{235}U metal were prepared having a thickness of 0.0002 in. and an area of 125 cm^2; these samples were for use in a fission chamber. For the first time, metal foils of isotopically enriched ^{236}U (99.8%) and ^{241}Pu (99.9%) were prepared; foils of ^{236}U were rolled to a thickness of 0.0001 in. and a circular area of 13 cm^2, and ^{241}Pu metal was prepared in rectangular foils of 12 cm^2 with a thickness of 0.0005 in.

RADIOISOTOPE TARGETS AND SOURCES

Radioisotope Facility

A new radioisotope containment area for the preparation of targets and sources was constructed and equipped. In designing this facility, emphasis was placed on handling heavy elements, principally the actinides and other alpha-emitting materials. Only weak beta- and/or gamma-emitting isotopes can be accommodated because of minimal shielding provided on the containment glove boxes surrounding fabrication equipment. Preparative techniques in this area are limited to vacuum evaporation-condensation, rolling, tritium sorption, and some electrodeposition; TIG welding equipment for source encapsulation is also included.

Two rolling mills in this area are enclosed in glove boxes in which purified argon atmospheres can be maintained. One mill is an 8-in., 4-high 2-high convertible Stanat machine capable of exerting 100,000-lb pressure on the rolls, while the other is a 2-in., 2-high chain-driven mill suitable only for relatively soft materials in small quantities. Argon-atmosphere annealing furnaces are enclosed within the glove box of the Stanat mill.

Glove box enclosures (steel) for the various vacuum evaporation systems are designed to handle only one element (including all isotopes); in this manner cross-contamination between the alpha- and beta-emitting heavy element isotopes is limited to only the small amount originating from earlier processing of the same element. Usually cross-contamination is not detectable, but it has never exceeded 0.01%. To reduce the frequency of removal or input of materials to any given glove box from the outside, all evaporator-containing enclosures are attached to an electric conveyor system through which materials can be transported to a central containment zone. Entrance to the conveyor from any glove box is through a normally sealed door which prevents escape of material to other boxes in the complex. The central box is used for preliminary packaging of materials, storing of isotopes, weighing of samples, pressing of powder compacts, and the initial cleanup of packaged samples. At present, evaporation facilities are available for Th, U, Pu, Np, Am, Cm, and Cf isotopes.

Californium-252 electrodeposition and/or molecular plating equipment is enclosed in a single isolated containment zone. This system can handle 10 μg of californium when necessary, but usually <5 μg is used at any one time. No other heavy elements are formed into targets or sources by electrodeposition since vacuum evaporation methods have proven to be superior for this purpose.

Two nonconnecting, isolated glove boxes have been equipped with vacuum evaporation equipment for forming special sources of ^{210}Po. Because of the special handling necessary with this isotope in elemental form, these systems could not be included in the complex previously described.

Each vacuum evaporator system used for heavy element target and source preparation is of the oil (DC-704) diffusion type which has an isolating high vacuum valve between the chamber and the 4-in. pump. Rough pumping of the systems is performed with a 1397-B Welch mechanical pump separately enclosed in a glove box. All exhaust gases from the pumps and glove boxes are vented to the cell ventilation system through absolute filters (0.3 μ); air intake to the containment enclosures is also filtered, and each box is maintained at a minimum negative pressure of 0.2 in. of water with respect to the atmosphere. Other than arrangements for back-filling the vacuum chambers (when necessary) with argon, no provision is provided for maintaining an argon atmosphere in the evaporation boxes. Each system is equipped with appropriate evaporation sources of the electron-bombardment and resistance-heating types.

Also included within the target facility are the various systems for preparation of tritium-containing targets of all sizes from <1.25 up to 22 in. in diameter. Two vacuum evaporation systems and two tritium sorption systems (glove boxed) are integral parts of this operation.

Considerable emphasis has been placed on quality control of radioisotope targets since the chemical purity of these materials is generally less than that of stable isotopes. A variety of counting devices and other metrological instruments have been included. Of particular importance are two low-geometry counting chambers using solid state detectors where count rates can be obtained from fabricated samples and semiquantitative identification of radioisotopic impurities can be made. Ion chambers have also been provided for very high-level samples such as ^{210}Po and ^{242}Cm deposits. Alpha, beta, and fission-fragment counting can all be performed on an absolute basis.

Targets and Sources Produced by Vacuum Evaporation Methods

More than 300 targets and sources have been prepared in the radioisotope facility by vacuum evaporation-condensation since operation began in March 1967. Many of these samples were of the "self-supporting" variety which were produced by evaporation of oxides or fluorides of the heavy element isotope on thin carbon substrates (substrate areal density 10-50 μg/cm^2). Most samples, however, were deposited films or coatings on heavy substrate materials of 0.001-in. thickness or more. Isotopes of Pb, Th, U, Np, Pu, Am, Cm, and Cf have been deposited in both self-supporting and supported forms. Since each target preparation involves a variety of routine operations described elsewhere[1], only a few unusual preparations will be discussed.

Fission Chamber Plates of PuO_2 on Aluminum

Accurate measurements of the fission cross section of ^{239}Pu are being performed by the Neutron Physics Division of ORNL using a fission chamber containing 21 plates of 0.005-in.-thick aluminum with a diameter of 3 in. which were coated with high assay ^{239}Pu. Nineteen plates were coated on both sides with PuO_2 by evaporation-condensation to an areal density of 1 mg/cm^2; the two terminal plates of the fission chamber were only coated on one side. The specified stacking of the plates in use required that the coated substrates be planar (no significant warpage).

Radiant heating of the substrates during PuO_2 deposition was found to be a major factor in deforming the substrates. However, unless the substrate temperature was raised to at least 350°C during deposition, poor film adherence resulted and this caused the deposit to crack and flake — especially on those plates which required coating on both sides. A suitable thermal pretreatment of each substrate was developed which eliminated these problems. Vacuum annealing of the substrates between planar copper blocks at 450°C proved to be satisfactory provided that a minimum temperature of 350°C was maintained on the substrates during PuO_2 vapor condensation and that the coated disk never exceeded the 450°C annealing temperature.

By using electron-bombardment heating of a tungsten cylindrical crucible (length, 1.25 in.; inside diameter, 0.20 in.), evaporation of the oxide at ~2400°C was performed without difficulty. By choosing appropriate evaporation geometry, radiant heating of the substrate produced the requisite surface temperature conditions, and simultaneous substrate rotation over the vapor source resulted in uniform deposition. About 15 g of the oxide was required to coat all disks. Each aluminum substrate was clamped between rigid stainless steel rings (designed to permit little flection of the aluminum) during oxide deposition. Tapering the inside edges of the stainless rings permitted high uniformity (93%) of deposition across the entire area.

Curium-244 Cross-Section Samples

Samples of ^{244}Cm were required for fission cross-section measurements in which the atom count of the curium was known to an accuracy of >95%. A vacuum-deposited curium oxide layer of ~50 μg/cm^2 areal density over a circular spot of 0.25 in. dia on a 0.423-in.-dia polished platinum disk was required.

Preparation of such samples with high-purity materials would have been relatively straightforward using direct areal density measurements (assuming that the stoichiometry of the deposited compound was also known), except that the available curium oxide was impure and the true nature of a vacuum evaporated-condensed deposit was difficult to define without extensive chemical and/or spectrographic analyses. Initial attempts to produce a variety of samples ranging in areal density from 10 to 100 μg/cm^2 (determined by direct weighing of the deposit) were unsuccessful. Subsequent alpha-counting of these samples at LASL proved that the quantity of curium present was less than expected by a factor of 200.

Low geometry alpha-counting equipment was designed to resolve this difficulty. By using an 87-cm distance between the radioisotope source and a silicon diffused-barrier detector, this counter system, together with a 256-channel analyzer and other electronic components, was able to resolve the two alpha-particle emission energies (5.80 and 5.76 Mev) of the ^{244}Cm species. Routine quantitative counting of sources could be attained, and the amount of curium present was then determined to a calculated accuracy of ±0.5%.

With this counting system, various curium targets of different thicknesses were analyzed. In all cases, the quantities of curium deposited, as determined by low-geometry counting, were less than those determined by direct weight measurement. Deviation factors of between 10 and 200 were observed with no apparent systematic variation. To obtain reproducible targets having the desired ^{244}Cm concentration, fractionation during vacuum evaporation was necessary. During initial heating of the evaporant in a tungsten tubular crucible, all vapors emitted before reaching the evaporation temperature of the CmO_2 (~2000°C) were condensed on a shield separating the vapor source and the target substrate. When the crucible was finally heated to the CmO_2 evaporation temperature, the condensing shield was removed, and evaporation-condensation of the residual material was allowed to proceed. Variations between mass measurements and ^{244}Cm assay by counting were found to be at least reasonably constant by this procedure; the difference factor was usually reduced to <10.

Three separate ^{244}Cm targets were prepared having a nominal areal density of 55 ± 1 μg/cm^2. Counting data used to determine the quantity of ^{244}Cm on each sample disk agreed with experimental data obtained at LASL to within ±1%. It was concluded that weight measurements of vapor-deposited radioisotope targets (heavy element materials) could not be relied upon to produce accurate atom-count information and only direct counting techniques would provide satisfactory analytical data unless pure starting materials of known composition were used.

Polonium-210 Coatings on Aerodynamic Forms

A process was developed to form adherent deposits of elemental ^{210}Po and several special forms were coated using this technique. Polonium, being a member of the same chemical family as tellurium, exhibits similar difficulties in forming adherent coatings because of its high vapor pressure at low temperatures. Since polonium-coated forms were to be used to study the effects of ionization for high velocity and static gas viscosities, good adherence of the coating was mandatory to create a constant alpha-emission intensity (the ionization source) and a low radioactive contamination potential, particularly at high gas velocities. As with tellurium, erbium metal subcoatings were employed to chemically bond the polonium to the substrates.

With polonium the additional problem of recoil sputtering was encountered because of the high energy (5.30 Mev) alpha emission. Some reduction in material loss by this mechanism was attained by overcoating the polonium film with metallic chromium (areal density ~100 μg/cm^2); the coating was made thin enough to permit nearly all the alpha particles to penetrate without significant energy attenuation. The thickness of the combined polonium coating film and the chromium overlayer was designed to permit maximum ionizing radiation (~35 mc/cm^2) to enter the gas phase surrounding the source.

The special forms coated were a 3-in.-dia aluminum disk (0.060-in. thick) and a 9°-right circular cone (3.5-in. length) made by machining pure titanium bar stock. In both cases the surfaces were precoated with chromium to form a tenacious bond between the aluminum or titanium substrate and the erbium metal layer. Each film of metal, including the ^{210}Po, was deposited by vacuum evaporation-condensation techniques. Chromium and erbium metals were evaporated by electron-bombardment heating a pellet of each of these materials, while polonium was evaporated from a tantalum tubular crucible heated by electrical resistance. Polonium as an electrodeposit on platinum gauze was used directly as the evaporation charge.

The most difficult problem was staying within the maximum atom density deviation of ±5% for the polonium deposit on the conical surface. Obtaining uniform films of chromium and erbium was straightforward since natural materials were being used and vapor source-to-substrate distances could be large. Only three times the required quantity of ^{210}Po was available; therefore, special evaporation geometries were studied and tested, using tellurium as the evaporant, before deposition of polonium was attempted. By calculation, a deviation in film thickness of ±7.5% from the mean value over the entire conical surface could be attained by using two tubular-crucible evaporation sources whose axes were perpendicular to the surface of the cone and which were located at the ends of the cone. However, testing of the system was required to determine the correct distance between the vapor sources and the conical surface.

The titanium conical substrates are illustrated in Fig. 14 with a segmented solid cone used to determine (by weight measurement) the areal density of tellurium films deposited at various distances from the vapor source. During evaporation, the cones were rotated at ~100 rpm in such a position that the surface was always parallel to a line connecting the two vapor sources. Probably as a result of deviation of the axes of the evaporation crucibles from the perpendicular, measured uniformity of mass distribution was found to be better than calculated — ±5% from the mean value.

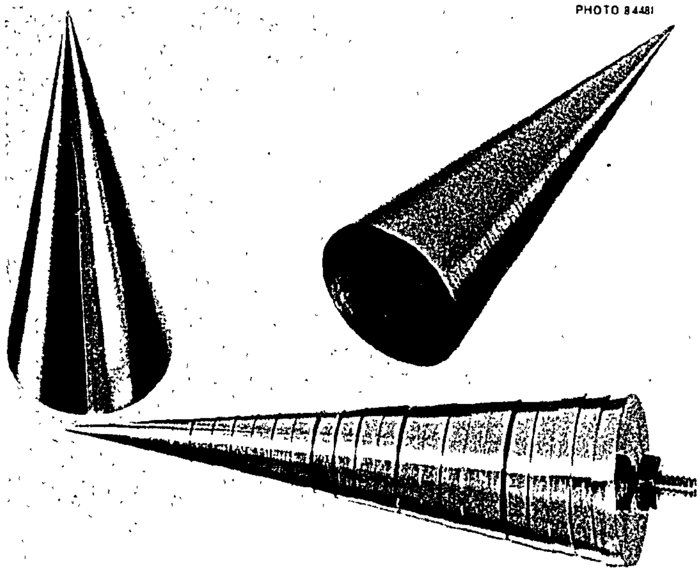

Fig. 14. Right-Circular Titanium Cones (9°) to be Used in Aerodynamic Studies. These cones were coated with chromium and ^{210}Po. The segmented cone was used to determine coating uniformity and evaporation geometry by direct mass measurement.

The alpha emission from one cone (coated with the various materials but prior to application of the chromium overcoating) was 18 mc/cm^2. The chromium overcoating was found to limit the dispersion of contamination to acceptable levels even when the sample was subjected to air velocities approaching Mach 3. The most active specimen of the several disk samples had an alpha emission from the surface of 38 mCi/cm^2. As with the conical sample, only minor contamination was observed after using the disk in an oscillating gas viscometer for a period of 14 days.

SPECIAL PROJECTS

Targets for the High Energy Neutron Reaction Experiment (HENRE)

As previously reported,[1] Project HENRE required the production of a neutron field of ~10^{13} n/sec for four consecutive hours. By utilizing the ^{3}H(d,n)^{4}He reaction, such a field was considered feasible provided a tritium-containing target could be fabricated which could withstand an estimated continuous deuteron beam power of 75,000 w without massive deterioration. To produce such a target, a water-cooled copper substrate was designed having a 1000-cm^2 surface on which could be deposited a layer of erbium tritide. This tritium-containing compound was chosen because of its high thermodynamic stability and resistance to decomposition under deuteron bombardment.

The active target surface was formed by vacuum evaporation-condensation of several metal films onto the copper substrate. A "graded seal" was developed to gain maximum adherence of the erbium to the substrate and thus provide the best heat transfer properties. Each copper substrate was first coated with a film of chromium metal to an areal density of ~250 μg/cm^2 while the substrate was heated to ~350°C; onto this surface erbium metal was condensed in a layer having an areal density of 2-2.5 mg/cm^2. This composite structure was found to withstand repeated thermal cycling from room temperature to 450°C and formed an adherent layer even after sorption of tritium into the erbium film. Accelerator tests proved that this surface was capable of maintaining its integrity under bombardment at current densities of >75 w/cm^2.

Vacuum environment and mode of deposition of the various metal coatings were found to be the most definitive parameters determining the neutron-generating capability of the finished product. Vacuum levels of 10^{-7} to 10^{-8} torr were necessary and were achieved during evaporation-condensation by use of a 500-liter/sec ion pump and a titanium sublimation pump, the combined capacity of which was estimated to be 20,000 liters/sec. Evaporator design and structural characteristics have been reported previously.[1]

To gain maximum uniformity of the vapor-deposited layers, the target substrate, weighing more than 40 lb, was rotated at 100 rpm behind a mask so metal vapors were condensed over a semicircular area equal to half the total target area at any instant. By maintaining a substrate-to-vapor-source distance of 15 in., a uniformity in areal density of 93% was achieved over the entire 1000-cm^2 target area.

Preparation of one such target required the evaporation of ~7 g of chromium metal, evaporation of ~65 g of erbium metal while maintaining the rotating substrate at >350°C, and a total of 6 hr, not including the time consumed in reaching the appropriate substrate temperature and cooling after evaporation-condensation was complete. Electron-bombardment heating of cylindrical bars of the various metals was used exclusively. After cooling to ambient temperature, a thin coating of cadmium metal (vacuum evaporated from a resistance-heated source) was deposited over the active target surface. This coating was used to inhibit surface oxidation of the erbium film during transfer of the target to the tritium-sorption system.

Tritium sorption was performed in an ion-pumped, all metal system (see Figs. 15 and 16), where the substrate was heated, under ultrahigh vacuum, to ~450°C; the ultimate pressure attainable in the chamber was measured at 2 x 10^{-9} torr. Tritium gas was purified by passing it over hot uranium chips and then sorbing it on cold uranium in the form of uranium tritide. Three uranium traps were used simultaneously to maintain a maximum inventory of 250,000 Ci of tritium gas.

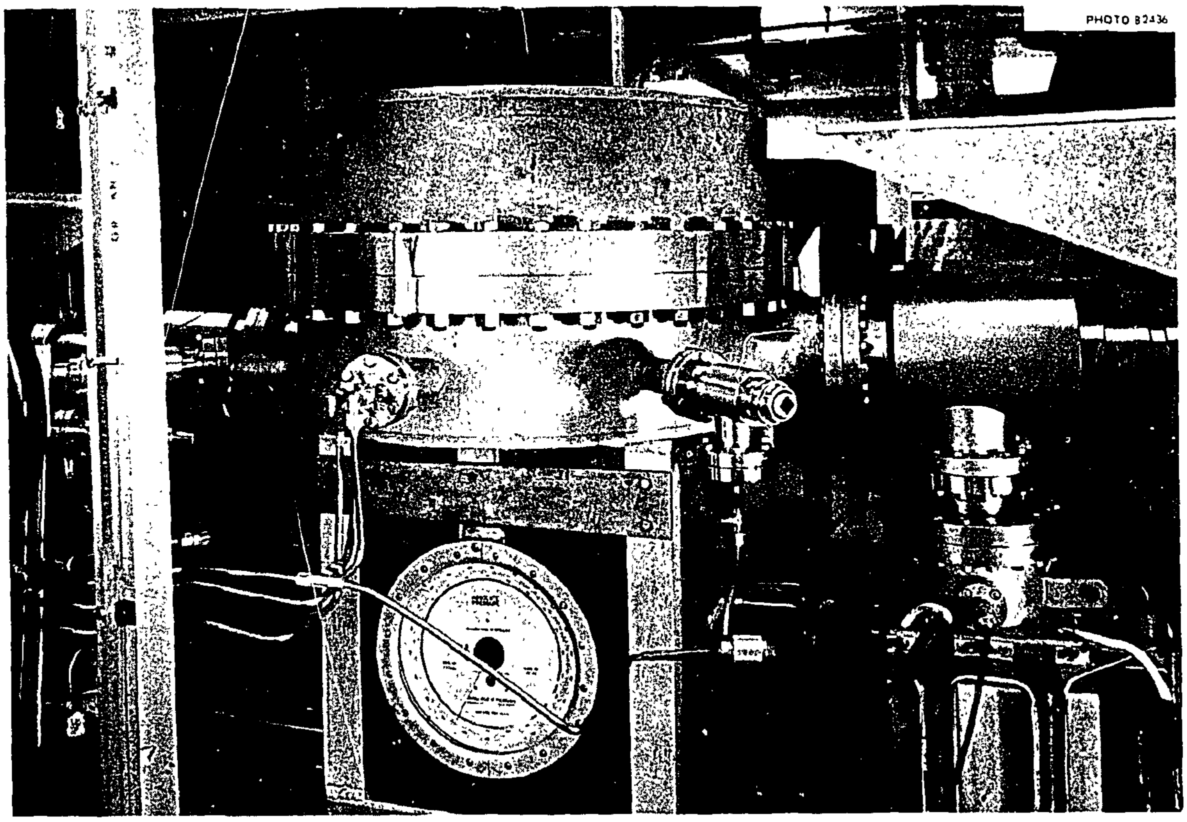

Fig. 15. Tritium Sorption System for Preparation of HENRE Targets. Entire system is stainless steel, has metal-to-metal seals, and is capable of being evacuated to 2 x 10^{-9} torr. The 200-liter/sec ion pump is shown in the lower right-hand corner.

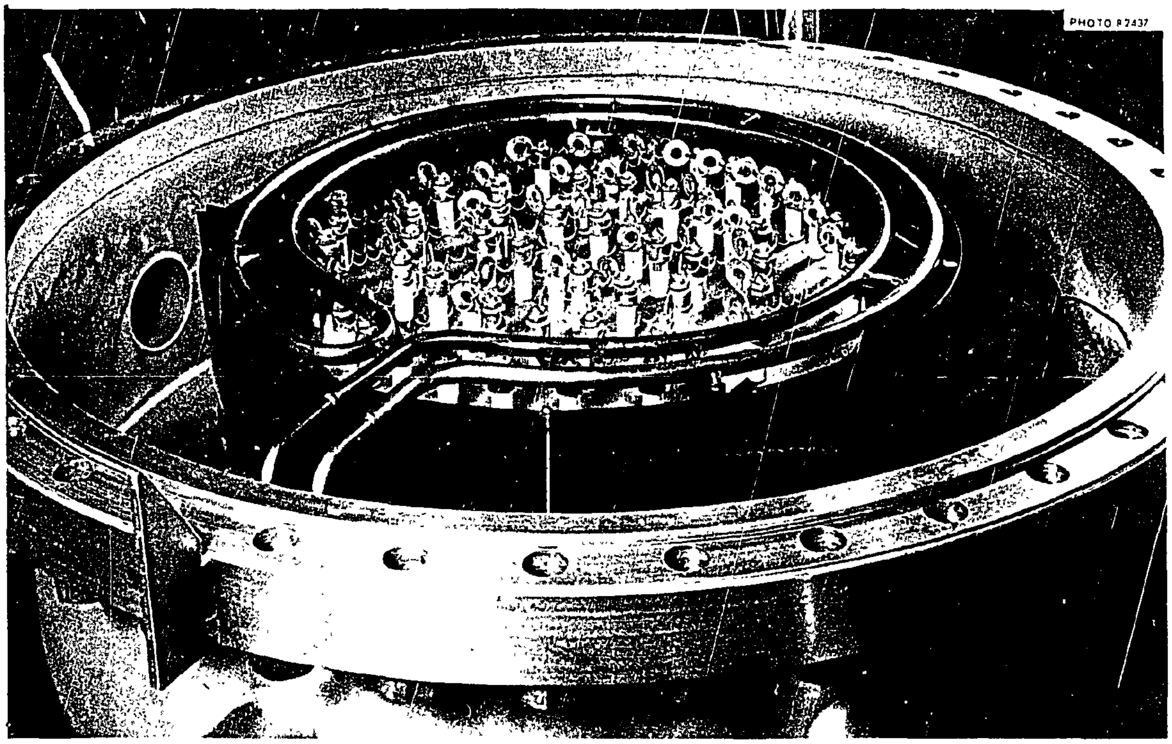

Fig. 16. Heater Assembly Interior to the Tritium Sorption System. The 40-lb copper substrate rests on the rings immediately above the coiled tungsten heater.

As can be seen in Fig. 16, the target substrate was placed on a metal ring immediately above a tungsten wire heater. This configuration permitted heating the entire substrate to any desired temperature up to 475°C. During the heating process, the cadmium overcoating was re-evaporated and condensed on the system walls, thus permitting contact between the tritium gas and the erbium metal sorption surface. When the target reached its maximum temperature and pressure equilibrium had been established (10^{-8} torr), the ion pump and titanium sublimator were isolated, and tritium gas was admitted to the system. Pressures of tritium between 180 and 450 torr were employed. After a "soak" period of 1 hr at the highest target temperature, the system was cooled to room temperature, the excess tritium gas was re-adsorbed on the uranium storage traps, and the system was brought to atmospheric pressure with argon. A plastic lid was bolted over the target surface (not in contact with the erbium tritide layer) to preserve an argon atmosphere over the active surface until it could be used.

Surfaces deposited on the target by this fabrication technique were usually of matte appearance and produced a bremsstrahlung level of 10 rem at 6-10 in. from the surface. Under deuteron bombardment, these targets generated a field of 5×10^{12} to 3×10^{13} n/sec for at least 4 hr (non-pulsed beam); several were bombarded for more than one period with average yields varying from 10^{12} to 10^{13} n/sec. A total of 40 such targets were prepared and at least 20 more are to be fabricated during the next year.

Preparation of High-Purity Calcium Disks

One hundred disks of high-purity normal-assay calcium metal with a 2 ± 0.001-in. dia and having a thickness of 0.010 ± 0.001 in. were prepared. In addition to the geometrical specifications, it was necessary to prepare these disks without adding impurities other than those normally encountered in the best available high-purity metal. For this purpose, high-quality calcium metal bars (1 in. in dia and 6 in. in length) were obtained which had a nominal chemical purity of 99.9%. Spark-source mass spectrometer analyses of these bars were made (at the ends and in the mid-section of each bar) to obtain an impurity profile of the starting material before and after cutting the bars into 1/8-in. thick disks. Each disk was then rolled using a 3-in. tool steel rolling mill in a purified argon atmosphere (containing <3 ppm oxygen, nitrogen, and water vapor). The subsequent sheets of calcium metal were cut in a precision punch, which is a scaled-up version of a 1-in.-dia punch capable of reproducing its diameter to ±50 μin. Measurements of all disks prepared indicated that the samples were dimensionally accurate to ±0.0005 in. in diameter and ±0.0005 in. in thickness. Spectrometer analysis of representative metal disks indicated no chemical impurities were added during this fabrication as far as could be detected (<1 ppm); the oxygen and nitrogen content of the samples remained unchanged. (It was noted that the oxygen and nitrogen content varied in the original material depending upon where the sample was taken from the bar stock.)

All calcium disks and associated scrap material were shipped under vacuum ($<10^{-6}$ torr) in stainless steel containers that had been prebaked under vacuum. Subsequent testing of this material indicated the metal was suitable for the specified purpose on all counts.

Ultrahigh Vacuum Electron Microscope

A standard Hitachi HU-11-B3, metallurgy-type electron microscope was purchased for use in examining thin isotope films prepared by the Isotopes Target Center. This instrument will be used in:

(1) Quality control of isotope foils being produced.

(2) Research and development of techniques for forming amorphous, polycrystalline, and/or single-crystal thin films.

Commercially available electron microscopes normally operate in the pressure region of 10^{-4} to 10^{-6} torr and are available only with oil-diffusion pumped columns. Greased O-ring seals for vacuum-atmosphere feed-throughs are used to transmit either rotary or linear motion (or both) from control rods outside the microscope to the various apertures and stages within the microscope. Subsequent contamination from hydrocarbon vapors originating in these greases, together with air in-leakage through the seals, results in only short-term examination of specimens and makes this type of construction unsuitable for present purposes.

The Center's Hitachi instrument is being modified to permit its operation at pressure levels of $\leq 10^{-8}$ torr. All feed-throughs will be replaced with "wobble-pin" bellows seals for transmission of rotary motion and shaft-type bellows seals for transmission of linear motion. If baked and out-gassed Viton elastomer O-rings and/or copper gaskets are used as static seals, little or no contamination will result, and ultrahigh vacuum can be achieved.

A new pumping system was also designed and constructed for the instrument. Three differential-sputter ion pumps (two with a capacity of 100 liters/sec and one at 20 liters/sec) are used to differentially evacuate the entire microscope column. Each 100-liter/sec ion pump is coupled with a 1000-liter/sec titanium sublimation pump, and all pumps are connected to the column through stainless steel manifolds (2- or 3-in. dia) fitted with stainless steel bellows to aid in alignment of the components and to dampen vibrations transmitted through the pump support rack. Figure 17 shows the evacuation system with its combined ion and titanium sublimation pumps.

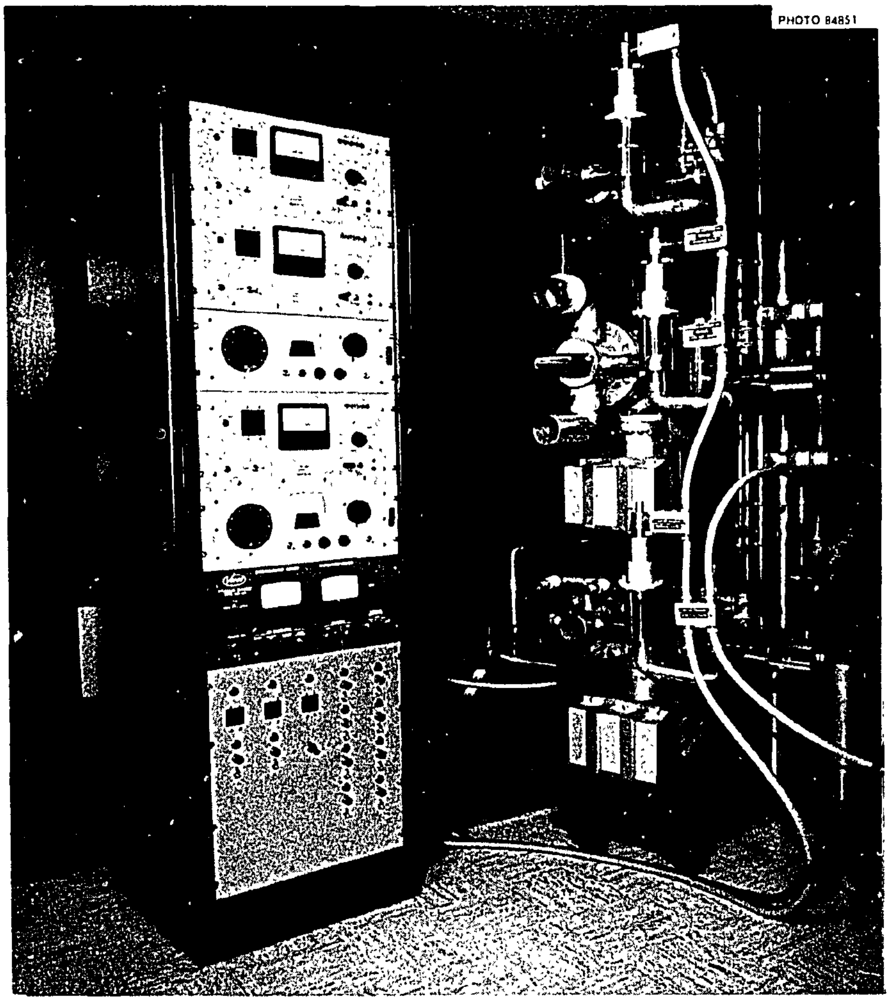

Fig. 17. Differential Ion Pumping System Developed for the Hitachi HU-11-B3 Electron Microscope. Two large molecular sieve trays are seen in the lower right-hand corner. These are used to evacuate the microscope to a pressure level at which the ion pumps can be started.

PUBLICATION

E. H. Kobisk, Isotope Targets for Nuclear Research, Nucleonics 24(8): 122-24 (August 1966).

PAPERS PRESENTED AT SCIENTIFIC AND TECHNICAL MEETINGS

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APPENDIX A

Target Preparations

Legend:
- a — evaporation
- b — rolling
- c — electrolytic deposition
- d — casting or pressing
- e — miscellaneous
- 1 — self-supporting
- 2 — metal backing
- 3 — thin carbon

Element	Range of thickness, $\mu g/cm^2$	Method of preparation	Backing	Form of deposit
Aluminum	5 to 12,000	a	2	Al
	150 up	a	1	Al
		d	1	Al
Americium	Undetermined	a		Am or oxide
Antimony	500 to 3000	a	1	Sb
Arsenic	10 to 200	a	2	As
Barium	10 to 1000	a	2	Salt
		d	1	Salt
	2000 up	a,b	1,2	Ba
Beryllium	Undetermined	a	1,2,3	Be
Bismuth	500 to 15,000	a	1	Bi
	10 to undetermined	a	2	Bi
Boron	20 to 250	a	1	B
	10 to 1000	a	2	B
Bromine	10 to 1000	a	2	Bromide
	5000 up	d	1	Bromide
Cadmium	10 to 200	a	2	Cd
	1000 up	b	1	Cd
		d	1	Cd
Calcium	10 to 1000	a	2	Ca
	700 up	b	1	Ca
		d	1	Ca or carbonate
Californium	100	c	2	Oxide
Carbon				
Normal	1.5 to 200	a	1	C
Isotopic	10 to 200	e	2	C

Target Preparations (continued)

Element	Range of thickness, $\mu g/cm^2$	Method of preparation	Backing	Form of deposit
Cerium	10 to undetermined	a	2	Ce oxide
Cesium	10 to 1000	a	2	Salt
Chlorine	10 to 1000	a	2	Salt
	5000 up	d	1	Salt
Chromium	500 to 12,000	a	1	Cr
	10 to undetermined	a	2	Cr
		d	1	Cr
Cobalt	1000 up	b	1	Co
	10 up	a	2	Co
	100 to 1000	a	1	Co
Copper	500 up	b	1	Cu
	10 to 1000	c	2	Cu
	200 up	a	1	Cu
		d	1	Cu
Curium	10 up	a	2,3	Oxide
Dysprosium	2000 to 25,000	b	1	Dy
	10 up	a	2	Dy
Erbium	10 to undetermined	a	2	Er
	2000 up	b	1	Er
Europium	10 to undetermined	a	2	Eu
	1000 up	b	1	Eu
Fluorine	25 to 500	a	2	Fluoride
Gadolinium	2000 to 25,000	b	1	Gd
	10 up	a	2	Gd
Gallium	10 to undetermined	a	2	Ga
	10 to 200	a	3	Ga_2O_3
		d	1	Ga_2O_3
Germanium	10 up	a	2	Ge
	300 to undetermined	a	1	Ge
Gold	10 up	a	2	Au
	Undetermined	a	1	Au
		d	1	Au

Target Preparations (continued)

Element	Range of thickness, $\mu g/cm^2$	Method of preparation	Backing	Form of deposit
Indium	5000 up	b	1	In
	10 up	a	2	In
		d	1	In
Iridium	10 to 100	a	2	Ir
Iron	500 up	b	1	Fe
	10 up	a	2	Fe
		d	1	Fe
Lanthanum	10 to undetermined	a	2	La
	2000 up	b	1	La
		d	1	La
Lead	8000 up	b	1	Pb
	500 to 8000	a	1	Pb
	10 to 1000	a	2	Pb
		d		Pb
Lithium	4000 to any cast size	b,d	1	Li
	10 to 500	a	3	LiH
	10 to 500	a	2	Li
Lutetium	10 to undetermined	a	2	Lu
Magnesium	150 to 1000	b	1	Mg
	Up to 20,000	a	1	Mg
		d	1	Mg
Manganese	300 to 1600	a	1	Mn
	10 to undetermined	a	2	Mn
Mercury	1000 to 4000	c	on Au	Hg
	Undetermined	a	2	Salt
Molybdenum	10 to 1000	a	2	Mo
	1000 up	b	1	Mo
Neodymium	10 to undetermined	a	2	Nd
	1000 to 15,000	b	1	Nd
Neptunium	10 up	a	2,3	Oxide
Nickel	500 up	b	1	Ni
	10 up	a	2	Ni
		d	1	Ni

Target Preparations (continued)

Element	Range of thickness, μg/cm^2	Method of preparation	Backing	Form of deposit
Niobium	10 up	a	2	Nb
	1000 up	b	1	Nb
Palladium	1000 up	b	1	Pd
	10 up	a	2	Pd
		d	1	Pd
Platinum	4000 up	b	1	Pt
	10 up	a	2	Pt
		d	1	Pt
Plutonium	10 up	a	2	Oxide
	5000 up	b	1	Pu (1% Al)
Potassium	7500 to any cast size	b,d	1	K
Promethium	10 up	a	2	Pm oxide
		a	2	Pm
Radium	10 up	a	2	Bromide
Samarium	1000 to undetermined	b	1	Sm
	10 up	a	2	Sm
Scandium	10 up	a	2	Sc
	1000 up	b	1	Sc
Selenium	300 to 5000	a	1	Se
	10 to 5000	a	2	Se
		d	1	Se
Silver	1000 up	b	1	Ag
	100 to 5000	a	1	Ag
	10 to 1000	a	2	Ag
		d	1	Ag
Silicon	10 to 1000	a	2	Si
	50 to 400	a	1	Si
	10 to 150	a	2,3	SiO_2
Sodium	7500 to any cast size	b,d	1	Na
Strontium	2000 up	b	1	Sr
Sulfur	5000 up	d	1	S
	10 to 100	a	2	S

Target Preparations (continued)

Element	Range of thickness, $\mu g/cm^2$	Method of preparation	Backing	Form of deposit
Tantalum	1000 up	b	1	Ta
	10 to 1000	a	2	Ta
Technetium	10 to 1000	a	2	Tc
Tellurium	10 to 1000	a	2,3	Te
		d	1	Te
Thallium	8000 up	b	1	Tl
	10 up	a	2	Tl
Thorium	4000 up	b	1	Th
	10 up	a	3	Th
Tin	4000 up	b	1	Sn
	10 to 1000	a	2	Sn
	500 to 4000	a	1	Sn
Titanium	500 up	b	1	Ti
	10 up	a	2	Ti
Tritium*	100 to 5000	e	2	Metal
Tungsten	In Cu matrix	b	2	W
	2000 up	d	1	W
Uranium	10 up	a	2	Oxide
	5000 up	b	1	U
Vanadium	1000 up	b	1	V
	10 up	a	2	V
Ytterbium	10 to undetermined	a	2	Yb
	1000 up	b	1	Yb
Yttrium	10 to undetermined	a	2	Y
	700 up	b	1	Y
Zinc	1000 up	b	1	Zn
	10 up	a	2	Zn
Zirconium	500 up	b	1	Zr
	10 to 5000	a	2	Zr

*Adsorbed on Zr, Y, Ti, or Er.

DISTRIBUTION

1-65. G. A. Kolstad, AEC, Washington
66. E. H. Kobisk
67. Laboratory Records, ORNL R.C.