ORNL - P - 1900
CONF - 651055 - 2

MASTER

S

PREPARATION OF ISOTOPE TARGETS BY ELECTRON-BOMBARDMENT TECHNIQUES*

E. H. Kobisk

Isotopes Development Center
Oak Ridge National Laboratory
Oak Ridge, TennesseeRELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

Problems encountered in the preparation of thin films of normal elements by vacuum evaporation-condensation techniques are significantly magnified when using isotopically enriched materials. These latter materials require maximum efficiency in evaporation and condensation and to achieve this end, the Oak Ridge National Laboratory Target Center developed an electron-bombardment vaporization source using high electron currents and low acceleration voltages. A minimum of ~20% and a maximum of 90% efficiency of vapor condensation have been obtained with this system. The vaporization source uses special crucibles to attain mechanical collimation, permitting the use of milligram quantities of evaporant in cases where previously several hundred milligrams had been required with other evaporation techniques. Materials having very high vaporization temperatures (~3000°C) have been evaporated successfully by this method, e.g., neptunium oxide, uranium oxide, plutonium oxide, and thorium oxide. Since the electron-bombardment heat source can be installed in all standard vacuum chambers at low cost and is suitable for use at pressures of 10^{-10} torr, it has proven a highly versatile tool.

The following characteristics of an evaporation system are desirable for preparing thin-film targets from isotopic materials

1. Variable evaporation temperatures between 300 and 3000°C.
2. Collimation of vapors for maximum recovery of starting material.
3. Compatibility of evaporant and crucible material to minimize loss of isotopes by chemical reaction or alloy formation.
4. Elimination or minimization of contaminants in the condensed films.
5. Maximum uniformity of film density.
6. Compatibility of the size of the evaporation system with the vacuum chamber.

Most of these criteria have been achieved with the vaporization system illustrated in Fig. 1.

From the Knudsen laws of gaseous flow through tubes, mechanical collimation of material can result if a tubular crucible is employed whose length is much greater than its internal diameter ($L/D \sim 10$). The evaporation crucible shown in Figs. 1 and 2 approximates these requirements.

*Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

However, choice of crucible materials is critical and was limited to those that would not alloy during fusion and vaporization of the specific isotope. The vaporization gun uses ring-type or hairpin-type electron-emission filaments, fabricated from 0.020- to 0.040-in.-dia thoriated tungsten wire and resistively heated with 25-100 amp ac to achieve the appropriate emission temperatures. Electrons drawn from the filament are directed toward the crucible by imposing a positive potential of 100 to 1000 volts dc between the crucible and the filament. Increased electron efficiency is achieved by surrounding the entire assembly with a 0.010-in.-thick tantalum box, electrically at earth potential, which acts as an electrostatic shield and increases the electron current to the crucible by ~30%. To raise the crucible temperature above 2000°C, an electron current of ~1.5 amp at a potential difference of ~800 volts between the filaments and the crucible was required.

To attain appropriate heat distribution and to minimize thermal losses, the crucible configuration illustrated in Fig. 2 was adopted. Crucibles were made from a tungsten rod 0.75 to 1.0 in. long and 0.25 to 0.125 in. in diameter. Heat conduction from the crucible was limited by a 0.040-in.-dia tungsten wire which also supported the crucible and resulted in a more uniform temperature distribution. Because of the high temperatures required, crucible materials were usually limited to tungsten, tantalum, and molybdenum. If crucible and evaporant were incompatible, ceramic liners (Fig. 3) could be used. One of the most versatile materials used for crucibles was found to be spectroscopic-grade graphite. Further collimation of vapors could be achieved using an effusion plug (shown at the right of Fig. 2), which proved very effective at short distances between the crucible and the substrate.

A variety of evaporation techniques was found possible using this versatile heat source. For example, as shown in Fig. 4 it was desired to coat the inside and outside surfaces of right circular cylinders with plutonium oxide. With resistance heating, this task would have been infeasible, particularly since good uniformity of the films on the surfaces was desired. However, with our electron-bombardment heating technique, successful deposition of the oxide over the inside and outside surfaces was achieved. At the bottom of Fig. 4 is shown the boat-type tungsten crucible used to contain the plutonium oxide while it was heated to ~2400°C by electron bombardment using a hairpin-type electron-emission filament. Each cylinder in the nest of cylinders was axially rotated at 100 rpm, with the evaporation crucible coincident with the axis of rotation of the cylinder. A second evaporation source of similar design was used to evaporate material onto the outer surface of each cylinder independently. The tungsten crucible was ~2 in. long and grooved down the center to allow even distribution of plutonium oxide powder before evaporation. Evaporation onto the outside surface of a cylinder is illustrated in Fig. 5.

Figure 6 illustrates the glove box housing and Pyrex glass "T" evaporator system designed for general use with radioisotopes. The electron-bombardment vaporization source is located centrally in the photograph. This particular system was used to deposit ^{244}Cm on the end of a 0.063-in.-dia copper wire.

The versatility of this technique is further illustrated by the fact that 0.2 μg of ^{252}Cf was evaporated with subsequent condensation of 50% of the starting material on the substrate. Of course, vaporization-condensation efficiencies of this high magnitude are not ordinarily achieved, particularly when good uniformity of the deposited material is desired over a large substrate.

One distinct advantage of this system is the low voltage employed to accelerate electrons toward the crucible. In most commercial systems, voltages of the order of 5000 volts dc are used. In practice, commercial electron guns cause electrical breakdown or glow discharge in the vacuum system following a burst of gas from the evaporant because of these high acceleration voltages. With the electron-bombardment system developed at the Target Center, it has been observed that discharge was avoided until pressures of the order of 1×10^{-3} torr were attained. Furthermore, because of the filament and crucible geometry, a rather high degree of ionization occurs in the evaporating material causing it to glow visibly, thus allowing control by sight of the evaporation. In the future it is hoped that, by redesign of the heat source, electrostatic and/or magnetic collimation of the vapors can be realized. It is conceivable that this technique can attain enhanced uniformity of deposition by sweeping the ionized vapor beam over the surface of the substrate rather than having it focused at one point and mechanically manipulating the substrate to gain good uniformity.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights, or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor

12/28/65

FIGURE LIST

Fig. 1. Electron-Bombardment Heat Source Used for Evaporation from a Tubular Crucible.

Fig. 2. Tungsten Evaporation Crucible: Components and Assembly.

Fig. 3. Crucible Liners That Have Been Used in Tantalum or Tungsten Electron Bombardment (left to right: BeO , ZrO_2 , and BN).

Fig. 4. Slotted Tungsten Crucible and Electron-Emission Filament (bottom) Used to Coat Fission Chamber Cylinders with $^{239}\text{PuO}_2$. (Fission chamber assembly is shown in the upper portion of the photograph)

Fig. 5. Evaporation of $^{239}\text{PuO}_2$ onto Fission Chamber Cylinder in a Pyrex Glass Cross Evaporator. Cylinder was rotated at 100 rpm above the vapor source.

Fig. 6. Glove Box Enclosed Evaporation System with Electron-Bombardment Heat Source in Place. This particular unit was used to evaporate ^{244}Cm as oxide onto a 0.600-in.-dia copper disk (as shown over the crucible).

Fig 1 Photo 66634

Fig 2 Plt 6663³

Fig 3 Plate 64670

Fig. 4
Photo 67322

Fig. 5 Photo 67320

Fig 6 Plt 66635

ORNL - AEC - OFFICIAL

ORNL - AEC - OFFICIAL

ORNL - AEC - OFF

ORNL - AEC - OFFICIAL