ISOTOPE TARGETS PREPARED BY VAPOR DEPOSITION

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ABSTRACT

As the state of the art for producing thin films has developed, the parameters set by the experimenters have in turn become more stringent. Often these targets must be of high purity, extremely thin, self-supporting, and of accurately determined thickness. In addition, the target material may be an expensive isotope which must be reduced to elemental form at the time of vapor deposition. Methods of producing some of these targets will be discussed with such examples as calcium, magnesium and boron.

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History

In 1930, Cartwright¹⁾ reported a method for making thin films. By cathode sputtering he produced mirrors of high reflectivity on glass surfaces. The mirror was found to be more perfect and rugged if the glass was first heated to 200°C in a vacuum and then the material sputtered on to it. The apparatus which he used is shown in Fig. 1. The most successful mirrors were those made from platinum or gold.

In 1931, Cartwright and Strong²⁾ reported a new method for depositing "most metals and some non-metals on various surfaces: which is what we now call resistive evaporations. Their system, shown in Fig. 2, used coils or crucibles as heaters for the evaporant. A vacuum of 10^{-4} torr was obtained by an oil pump for roughing out the system and activated charcoal fingers cooled to liquid nitrogen temperature for the high vacuum necessary for the evaporation.

Since that time, several new methods have been developed for producing thin films. These include resistive evaporations, electron beam systems, electron heating, sputtering equipment, and several others.

Parameters of Some Difficult to Produce Thin Films

With the development of new accelerators, refined technology for doing experiments, and new types of experiments, it now often becomes necessary to produce a thin film or "target" having some or all of the following characteristics:

1. High purity
2. Expensive separated isotope
3. Very thin
4. Self-supporting
5. Targets which oxidize readily in air

To produce a target of high purity, one must use pure source material, have a clean evaporator system, and reduce contaminants from the pumping system to a minimum. Thomas et al.³⁾ have discussed the value of having pure source material as is graphically shown in Fig. 3. They have also discussed the need to reduce the contaminants from the pumping system and to evaporate the source material in as good a vacuum as possible, preferably 10^{-6} torr or better. By using a cryo-pump to produce the high vacuum, the carbon, nitrogen, and oxygen contaminants are greatly reduced.

In many cases, the experimenter needs multiple targets of different thicknesses. At Argonne, Evans⁴⁾ has designed the system shown in Fig. 4 which has two tiers of target holders, thus allowing the production of different target thicknesses from a single evaporation. In addition, any individual target may be selected and removed from the evaporator by means of a small vacuum chamber and transferred to a vacuum storage chamber without the target being exposed to the atmosphere. Targets have been stored successfully in the vacuum storage chamber, previously described by Worthington⁵⁾, for over five years. Twenty-one targets can be produced from one evaporation, thus greatly reducing the cost per target.

Because of the high cost of some separated isotopes, it is necessary to carefully plan the method for producing targets from these materials. It is best to first test the procedure using normal material, before the actual isotope

is used. Often the isotope must be reduced to metallic form either before or at the time of evaporation. A useful reference for these procedures is given by Haegney & Haegney⁶⁾, who give information on reduction techniques, reducing agents used, etc.

We will describe in the following example the procedures used for making targets having some or all of the characteristics mentioned above.

Boron

A number of workers have reported on methods for preparing self-supporting boron foils⁷⁻¹⁶⁾. The technique described here has been given elsewhere by Thomas¹⁷⁾; some of the more significant features of that paper will be mentioned here.

Using an evaporation system separate from that for the boron, $50 \mu\text{g}/\text{cm}^2$ of NaCl is deposited on 2.5×7.5 cm microscope slides. These slides are then placed in a clean evaporation system which uses an electron beam to first melt and then evaporate the source. A vacuum pressure of 10^{-6} torr or better is used to improve the quality of the evaporation as well as minimize the oxygen and nitrogen contaminants. The substrates are heated to 175°C as suggested by Arnison and Gilmore¹⁴⁾, to reduce stresses in the boron so that a continuous film can easily be removed from the glass slide.

High purity crystalline boron obtained from Eagle Picher¹⁸⁾, having pieces #4 mesh in size is used as source material. This is a necessity for a successful evaporation. The boron is heated to 800°C by an electron beam to remove any H_2 residue and then further heated very slowly until a small puddle appears on its top. The puddle size is then held constant by controlling the temperature. Again, it is important to note that at this point the crystal

would explode if impure materials were used; as the impurities evaporate first and the vapor pressures inside the boron from these impurities would cause the source to explode. However, with a high purity material, the rate of evaporation can be maintained at $\sim 1/\mu\text{g}/\text{cm}^2/\text{min.}$ with relatively small input current from the e-gun and with no sputtering from the source.

After the evaporation is completed, the substrate heaters are turned off, the e-gun cooled, the slides removed from the system, and boron is easily floated off in water.

Calcium

Some of the techniques used in this method are those suggested by Ellsworth¹⁹⁾. Self-supporting foils of $150 \mu\text{g}/\text{cm}^2$ or less can be produced by the resistive evaporation method. A low humidity room atmosphere is desirable to increase the chance of success.

Brillantshine²⁰⁾ is used as a release agent. It is first rubbed onto a 2.5×7.5 glass slide for about 30 seconds using a lint-free tissue²¹⁾. After the coating has dried (about 5 minutes), it is rubbed a second time and then additional Brilliantshine lightly added to produce a continuous coat of material. After a period of drying, the surface is dusted with a camel's hair brush, making sure that all particles visible under an illuminated magnifier are removed. The slides are then ready for use.

The following reactions occur when producing a calcium target from calcium carbonate source material:

Stoichiometric proportions are used with the weight ratio of 1 part of CaCO_3 to 1.8 parts of Zr. Other workers have their favorite reducing agents such as La, Th, or Y, but we have obtained the best results with Zr. It is quite stable and has no radioactive residue.

Procedures used for the evaporation are:

1. Heat CaCO_3 in air to 1000°C to reduce to CaO.
2. Mix one part CaO and 1.8 parts fresh filings of crystal bar Zr chips.
3. Press into a pellet.
4. Place pellet into closed Ta boat (6.4 mm. d., 2.56 mm. wall thickness, 0.82 mm. d. hole for evaporant).
5. Use source to target distance of 7.5 cm.

It is best to evaporate in short bursts so that the substrate does not overheat.

A thickness monitor²²⁾ is used to measure the thickness of the evaporation.

Generally it is found that under these conditions a 100 mg. sample of CaCO_3 produces a layer of Ca metal $\sim 200 \mu\text{g}/\text{cm}^2$ thick.

After the evaporation, the target must be protected from oxidation.

Argon gas is slowly admitted to the bell jar until atmospheric pressure is reached. With the operator wearing thin cloth gloves, the slides are immediately transferred to a glove box containing argon gas. A single edge razor blade is cleaned with alcohol and, in the manner shown in Fig. 5, the calcium foil is carefully removed from the slide using the blade as a draw knife. Pressure should be exerted against the slide, and the blade should be moved rather slowly. It is advisable to scribe the foil before cutting, so that the desired size of target can be removed from the slide. Using tweezers, the foils are then placed on target frames. A very thin coating of vacuum grease on the frame has been found to be sufficient to hold the target on the frame.

Magnesium

The technique for producing magnesium targets is quite similar to that for calcium. There are, however, a few differences which should be pointed out.

The following reaction occurs when producing magnesium foils from MgO and using zirconium as a reducing agent as suggested by Takayanagi et al.²⁴⁾:

Twice the stoichiometric ratio of Zr works quite well or by weight 1 part of MgO to 2.3 parts of Zr chips. The yield is only about 50% of that obtained for calcium. A mixture of 85 mg. of MgO and 200 mg Zr produces ~150-200 $\mu\text{g}/\text{cm}^2$ of Mg. The other procedures are the same as for calcium.

The oxidization problem for Mg targets compared to that for calcium is minimal. However, several groups of experimenters have found that oxidization of the magnesium presents a problem in their particular experiment and as a result we find it desirable in these cases to store the targets in the vacuum storage chamber after preparation.

Magnesium foils are much easier to remove from the frames than calcium and they seem to be tougher. In addition, Gursky²⁵⁾ has developed a method for producing self-supporting magnesium films having a thickness of 75 $\mu\text{g}/\text{cm}^2$ or less. Magnesium is first evaporated on to the slide in the usual manner. The slide is then dipped in a formvar solution, the foil is stripped from the slide, mounted on a frame, and then the formvar is removed from the film with ethylene dichloride.

Karasek²⁶⁾ has also reported on a method for producing thin magnesium foils. Magnesium of ~200 $\mu\text{g}/\text{cm}^2$ is first evaporated onto a glass slide and then removed as a foil with a razor blade. This foil can then be rolled to thicknesses down to 50 $\mu\text{g}/\text{cm}^2$.

Conclusions

If history can be our guide, the art of producing vacuum vapor deposition films promises many exciting new techniques yet to be discovered and phenomena to be explained. The future parameters for an evaporator system may include a very clean oil-free system, ultimate evaporation pressures of at least 10^{-8} torr, targets with fewer impurities, thinner, thicker, etc.

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Figure Captions

- Figure 1. System used by Cartwright¹⁾ in 1930 for sputtering thin films.
- Figure 2. System used by Cartwright and Strong²⁾ in 1931 for resistive evaporation of thin films.
- Figure 3. Boron spectra obtained from two different targets - one using source material of high purity boron and other using less pure boron.
- Figure 4. System for producing many targets from one evaporation and transferring them to storage chamber without exposure to the atmosphere.
- Figure 5. Method for removing thin films from substrate using a razor blade.

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<u>RATIO</u>	<u>OLD</u>	<u>NEW</u>
$\frac{\text{C}}{\text{B}}$	0.40	0.51
$\frac{\text{O}_2}{\text{B}}$	0.45	0.36

Revised

ANL-P-15,977