Preparation of Calcium-Separated Isotope Targets Using Small Samples

By

G. E. Thomas

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Argonne National Laboratory, Argonne, Illinois 60439

Abstract

Targets are routinely evaporated using a few milligram quantities of separated isotopes of calcium with reducing agents. The source to target distance is 3.0 cm with the substrate, if necessary, as thin as 15 $\mu g/cm^2$ carbon or 100 $\mu g/cm^2$ of gold. A tantalum closed boat, heat shield, and special collimator system are used.

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Introduction

Many of the experiments, particularly at the tandem Van de Graaff and Dynamitron accelerators at Argonne National Laboratory, use calcium targets of either natural-abundance material or separated isotopes. The thickness of the targets which must be deposited varies from 5 $\mu g/cm^2$ to 500 $\mu g/cm^2$. Because of the high cost of some of the separated isotopes, it is desirable to use a minimum of such material. Our methods have evolved from our first initial efforts which depended largely on black magic and luck -- and for which the quality of the targets obtained varied greatly from one evaporation to the next -- to the procedure used today which has a certain amount of logical basis and yields reproducible results rather routinely. However, there are still times when, because of such things as "a humid or cloudy day", it is difficult to evaporate a good target.

Our needs are somewhat different from that described by Stinson[1)] for the evaporation of Calcium metal. A new evaporation system has been purchased with the purpose of producing better targets faster. It has a high pumping speed with a good ultimate vacuum but it was soon discovered that commercial evaporation systems often need modifications before they are suitable for use with thin, fragile targets. For example, when we first loaded the evaporator with several targets having thin carbon substrates and turned on the roughing pump switch, the valve opened with a POOMPF and all the carbon substrates disappeared. Some adjustments had to be made to the automatic valving system.

Procedure

If in a calcium evaporation one starts with the compound calcium carbonate, there are two basic reactions which occur in our method of evaporating a target. They are the following:

1. $CaCO_3 + \text{heat} \rightarrow CaO + CO_2$
2. $2CaO + Zr \rightarrow ZrO_2 + 2Ca$ (Zr is an example of several reducing agents that can be used.)

It doesn't really make much difference whether one starts with the carbonate or oxide. We have converted the carbonate to the oxide by heating it in a platinum boat to 1000° (Ref. 2) in an air atmosphere but it works just as well to heat it under vacuum at the beginning of the evaporation.

In our procedure, the calcium carbonate is first mixed with a reducing agent and then pressed into a pellet using the die shown in Fig. 1. The pellet shown in the figure will produce a 100 $\mu g/cm^2$ target. The close contact of the materials in the pellet causes more complete and easier reduction. Also, after evaporation the pellet can again be weighed to determine the completeness of evaporation.

The pellet is then placed in a closed tantalum boat made from a piece of 1/4' dia. tantalum tubing. A 1 mm dia. hole is drilled in the center of the tube for evaporation and the ends are pinched shut. This is shown in Fig. 2 with the electrodes on either side and the thickness monitor above. With a closed boat such as this, one can better confine the heat and in addition the target yield is a factor of two higher than one would expect from a solid-geometry calculation using an open boat. The heat shield shown in Fig. 3 is then mounted above the boat. It is made from a piece of tungsten 1/32" thick with a 1/8" dia. hole which will allow evaporation of the calcium and at the same time give a certain amount of protection to the substrate from the heat generated.

The collimator shown in Fig. 4 is next placed over the heat shield and carefully aligned with the center of the target frame. It is a combination of stainless steel sheet and tubing to produce a collimator 3/8" I. D. and 3/16" high. If one has a 1/2" dia. hole in the target frame and a 3/8" dia. target, there is a carbon substrate ring between the frame and the target which will flex and reduce the chance of breaking the target substrate during evaporation or when it is exposed to the accelerator beam.[3)] Also, if the target is evaporated over the whole substrate, the target and substrate have a tendency to roll off of the target frame during evaporation.

We are now ready to start the evaporation. The thickness monitor is placed about 5 cm above the boat so that one can tell from the monitor when the calcium just starts to evaporate. The temperature is then raised until the vacuum gets poorer which is the signal that the calcium carbonate is reducing to calcium oxide. After the vacuum recovers, the temperature is raised until the monitor shows that the evaporation is just starting. The target frame is then inserted into the position shown in Fig. 5 and the temperature raised to that point which has been previously shown, by having no target in the beam and evaporating onto the monitor only, will completely evaporate the calcium. Since the target is only 3.0 cm from the boat it is important to raise the temperature only high enough for evaporation to occur in order to reduce the possibility of excess heat destroying the substrate. The target frame is left in the beam for ~25 seconds, or longer, depending on the evaporation thickness needed, and then removed immediately. If left longer the target substrate may be destroyed or due to excess heat some of the calcium may re-evaporate off of the carbon substrate.

The target is then transferred in a vacuum transfer system to a storage chamber having a vacuum of 10^{-6} Torr. Calcium targets of this thickness must be kept in a vacuum at all times to retain their quality.

Analysis

Several measurements have been made with an optical pyrometer to determine the temperature at which calcium evaporations occur using various reducing agents. The results of these measurements are shown on the graph in Fig. 6. First, we find that at 10^{-6} Torr, $CaCO_3$ reduces to CaO at a temperature of 750°C—900°C. The actual evaporation temperature, if one uses CaO with no reducing agent, is ~1700°C. With Zr as a reducing agent it is lowered to 1325°C and with Y to 1200°C. Since, with these small samples of expensive separated isotopes we are evaporating at a distance of only 3.5 cm, one can readily see that it is very important to use a reducing agent that will lower the evaporation temperature as much as possible and thus minimize the heat generated.

We have made a study with R. Ackerman4) to determine which reducing agents are best from the standpoint of free energy change. The results of his calculations are shown in Table I. For comparative examples, if one uses titanium as a reducing agent the free energy change is 33 kilocalories per mole, with zirconium it is 15, and for yttrium it is a low of -4.7. From our experience yttrium as a reducing agent does evaporate calcium at the lowest temperature of those reducing agents listed in the table. As a check to verify that calcium is actually evaporating when one uses yttrium as a reducing agent, we find that yttrium alone evaporates at a much higher temperature than calcium plus yttrium (~1500°C).

We do not find lanthanum to be a suitable reducing agent in our procedure because it actually melts before it reduces. This apparently causes sputtering, which destroys the carbon substrate before any calcium evaporates. This may be no problem if evaporating onto a thick substrate.

Acknowledgments

I would like to thank the many people not mentioned who have shared their techniques for calcium evaporation so that this method could be put together. Their contributions are greatly appreciated. I would also like to thank Dr. L. M. Bollinger, Director of the Physics Division, for his continued interest and support in developing better targets.

References

[*] Work supported by the U. S. Energy Research and Development Administration.

[1] J. D. Stinson, Proc. of the 1974 Ann. Conf. of Nucl. Target Dev. Soc., Chalk River Nuclear Laboratories.

[2] E. H. Kobisk, Oak Ridge National Laboratory, private communication.

[3] P. Maier-Komor, Technical University of Munich, private communication.

[4] R. J. Ackerman, Argonne National Laboratory, private communication.

Table I

Reducing Agents for CaO[*]

Reaction	Free Energy Change[a] (STD Conditions)	FEC per mole Ca
$Ti + 2CaO \rightarrow TiO_2 + 2Ca$	66	33
$Zr + 2CaO \rightarrow ZrO_2 + 2Ca$	31	15.5
$2La + 3CaO \rightarrow La_2O_3 + 3Ca$	13	4.3
$Th + 2CaO \rightarrow ThO_2 + 2Ca$	3	1.5
$2Y + 3CaO \rightarrow Y_2O_3 + 3Ca$	-14	-4.7

[*] Calculations made for Temp. at 1500°K.

[a] Kilo calories for equation as written. Values courtesy of R. J. Ackerman, Argonne National Laboratory.

Figure Captions

Fig. 1. Die used to press mixture of calcium and release agent into a pellet.

Fig. 2. Photograph showing closed tantalum boat and evaporation electrodes.

Fig. 3. Photograph showing heat shield over the closed boat.

Fig. 4. Photograph which illustrates the collimation system as well as the thickness monitor.

Fig. 5. Photograph of the target in position for evaporating.

Fig. 6. Graph of power output of system vs. temperature (degrees C) of closed boat and indicating evaporating temperatures of various materials used.

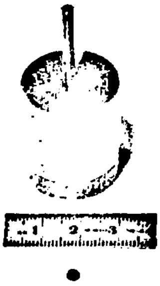

FIG. 1

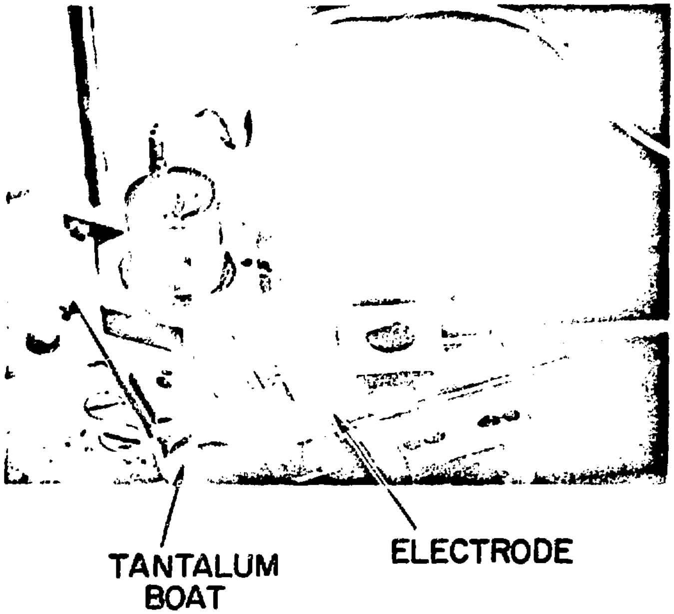

FIG. 2

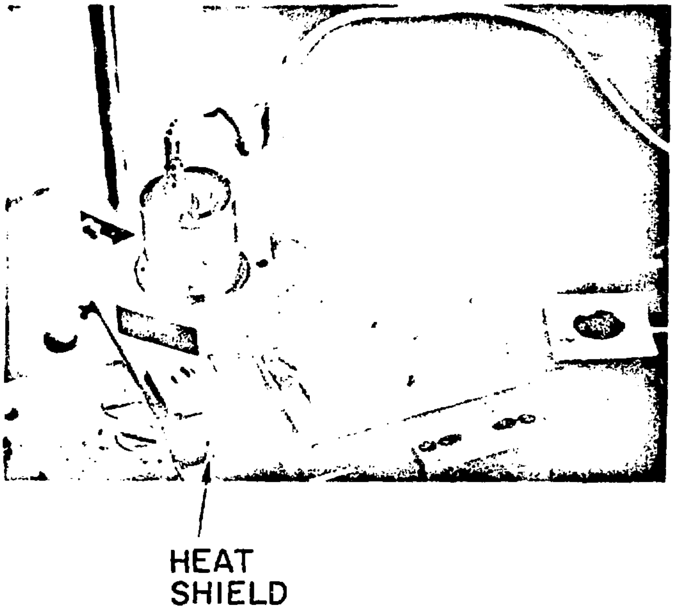

FIG. 3

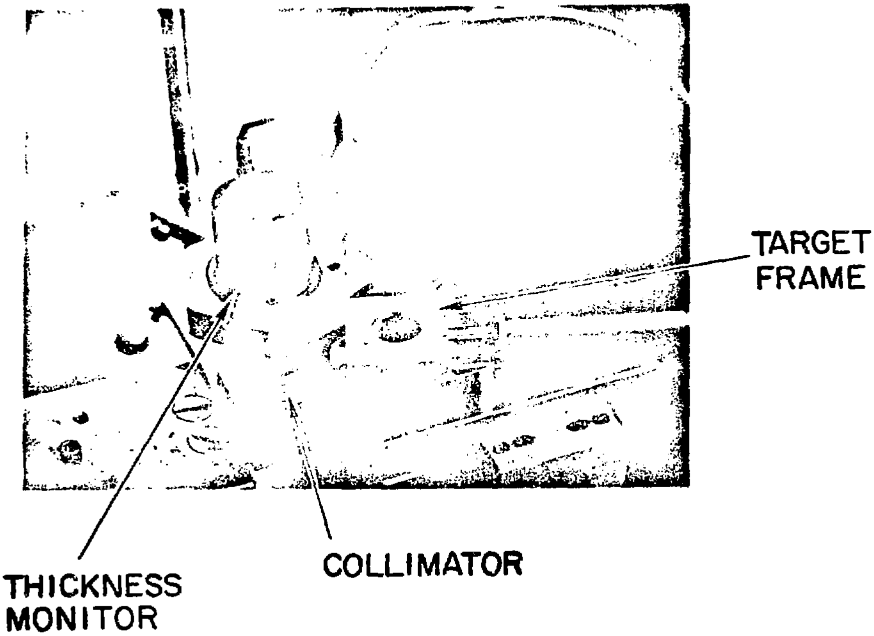
TARGET FRAME
COLLIMATOR
THICKNESS MONITOR

FIG. 4

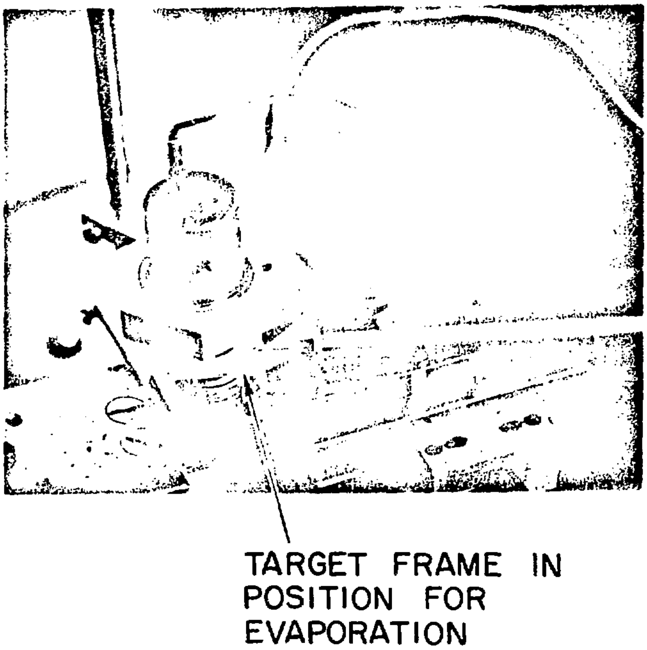
TARGET FRAME IN POSITION FOR EVAPORATION

FIG. 5

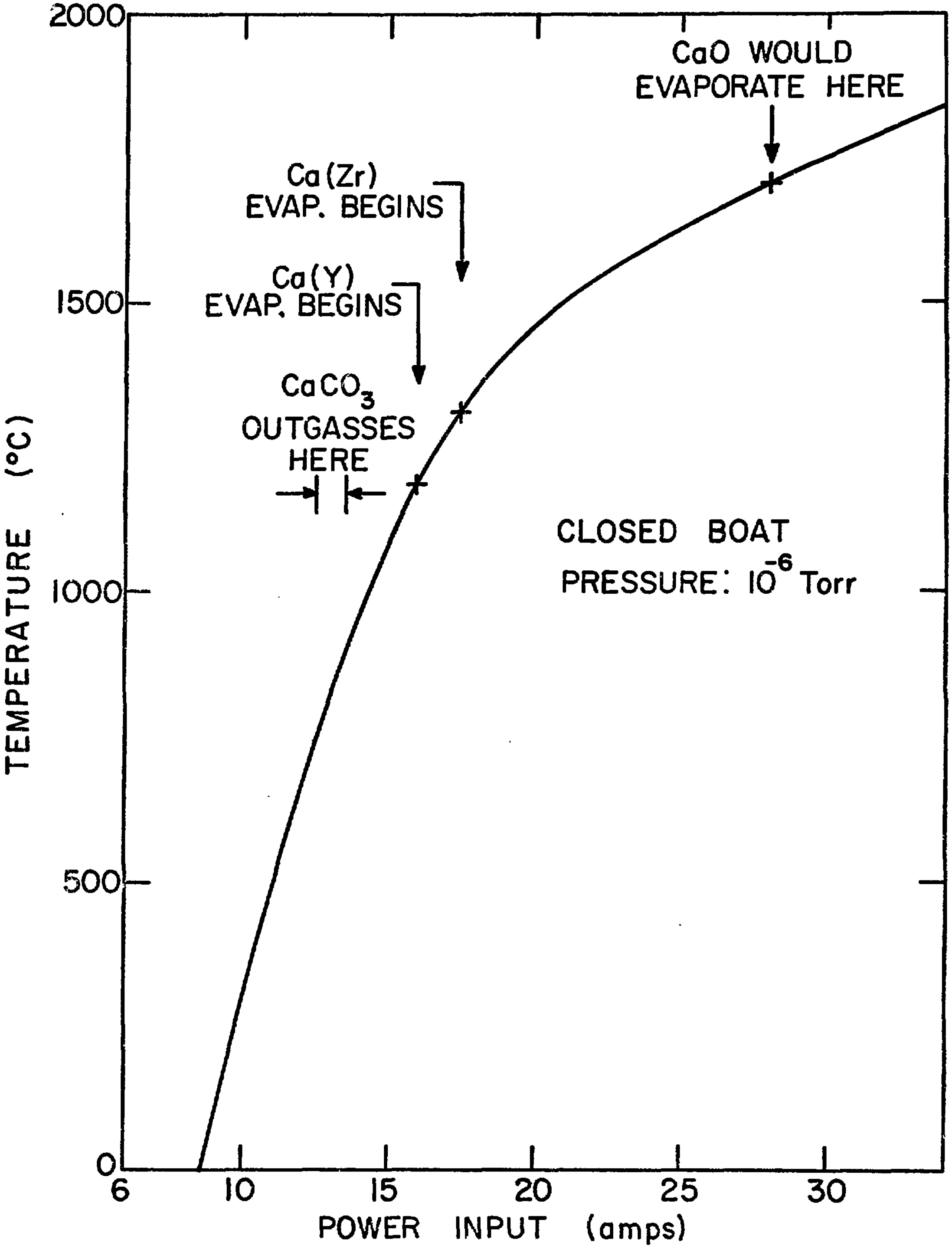
2000
1500
1000
500
0
6
10
15
20
25
30
TEMPERATURE (°C)
POWER INPUT (amps)
CaO WOULD EVAPORATE HERE
Ca(Zr) EVAP. BEGINS
Ca(Y) EVAP. BEGINS
CaCO3 OUTGASSES HERE
CLOSED BOAT
PRESSURE: 10^-6 Torr

FIG. 6