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THIN FILMS OBTAINED FROM MATERIALS OF HIGH EVAPORATION TEMPERATURES AT LOW PRESSURES

D. Pulino, G.M. Sipahi, G.M. Ukita,
T. Borello-Lewin, L.B. Horodynski-Matsushigue, J.L.M. Duarte,
W.G.P. Engel and J.C. de Abreu

Instituto de Física da Universidade de São Paulo
Caixa Postal 20516
01498 - São Paulo,SP

ABSTRACT

Recommendations for the fabrication of thin, clean and uniform films of materials of high evaporation temperatures at low pressures are presented. The convenience of the use of electron bombarded source techniques, with especially well defined focus, precautions to avoid overheating and the necessity of excellent cleaning are stressed. Contamination by higher vapor pressure substances and other known difficulties associated with the handling of materials at high temperatures were minimized. Good quality films of Zr, Mo and Ru, for use in nuclear physics experiments, were obtained.

1. INTRODUCTION

The restrictive conditions imposed by high resolution nuclear charged particle spectroscopy on thin films used as targets, led us to invest in techniques for the fabrication of clean, uniform layers (20 - 200 $\mu g/cm^2$) of the required materials. The isotopically enriched elements are normally high-cost substances of which only extremely small quantities (5 - 50 mg) are available and, to guarantee success, all steps of the method must be dominated. The technique is, therefore, developed with the substance in its natural, not enriched form, and followed by extensive training. This paper refers to recommendations which can be of more general interest in the handling of conducting materials of high evaporation temperatures at low pressures. They result from knowledge accumulated especially in controlling the set of procedures necessary to obtain good quality Mo targets [1] and complement and systematize previous experience [2] of the authors and other members of the Pelletron Laboratory. The evaporation temperatures of Mo, Zr and Ru, of which adequate films were obtained by the presently described method, are, as catalogued by a fabricant [3] at 10^{-5} Torr, all above 1860 ^{o}C.

2. EXPERIMENTAL METHOD

A standard EDWARDS evaporation system [4] (model E12E3), shown schematically in Fig. 1, was used. Numbers identify the most important components of the system. High vacuum is provided by an oil diffusion pump (backed by a mechanical pump) in a chamber limited by a pyrex glass bell jar, which allows visual accompanying of the process. The shutter can be commanded from the exterior and is important in the process.

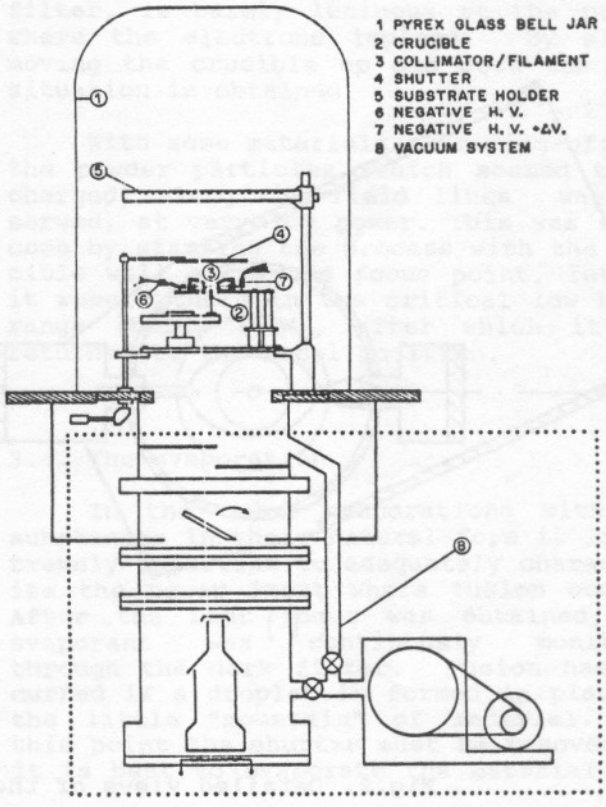

Fig.1. Scheme of the evaporator

Electron bombardment (E.B.) [5] was chosen as the heating method, since it optimizes the energy transfer to the material to be evaporated, in a controlled manner, detailed later. The evaporant is, thus, by far the hottest point of the system and contamination is minimized. The standard arrangement in use (E25016), is shown in Fig. 2. A negative high voltage (-H.V.) is applied to the collimator, which is of convenient form and shields the circular electron emitting filament of W or Ta. The filament is heated by a low potential difference (ΔV) with respect to -H.V. The crucible, of conducting material with high fusion temperatures, is fastened through a conducting support to the Cu base, which is connected to earth. The crucible is precisely situated at the axis of the collimator and may be moved in the vertical direction from the exterior of the chamber. This allows the adjustment of focus of the electrical field lines on the material to be evaporated, provided it is conducting.

The vapor of interest was deposited on thin carbon films, either still on their glass supports or already stretched on target frames, in which case spot targets could be obtained by the use of appropriate masks. Self-supported targets were obtained for Zr by a specially developed method [6] and its extension to Mo targets is presently being investigated.

3. RECOMMENDATIONS

3.1. Cleaning of the system

Thorough cleaning of the evaporator system was found extremely important in these high-temperature procedures to avoid contamination by previously evaporated lower-temperature materials and other adsorbed substances. All parts of the electron bombardment arrangement and its vicinity were disassembled and subjected to sand-jet and subsequent chemical cleaning. The vacuum bell jar and evaporator base were also adequately cleaned. A careful pump-down in all phases is important to establish a good base-vacuum ($\lesssim 5\ 10^{-6}$ Torr) in the system. To minimize contamination by the silicon of the pump-oil, seen on some targets, it was found important to always open the master-valve only after complete cool-down of the cold-trap and preferably to use a carbon oil.

Before depositing the evaporant in the crucible, a different one for each

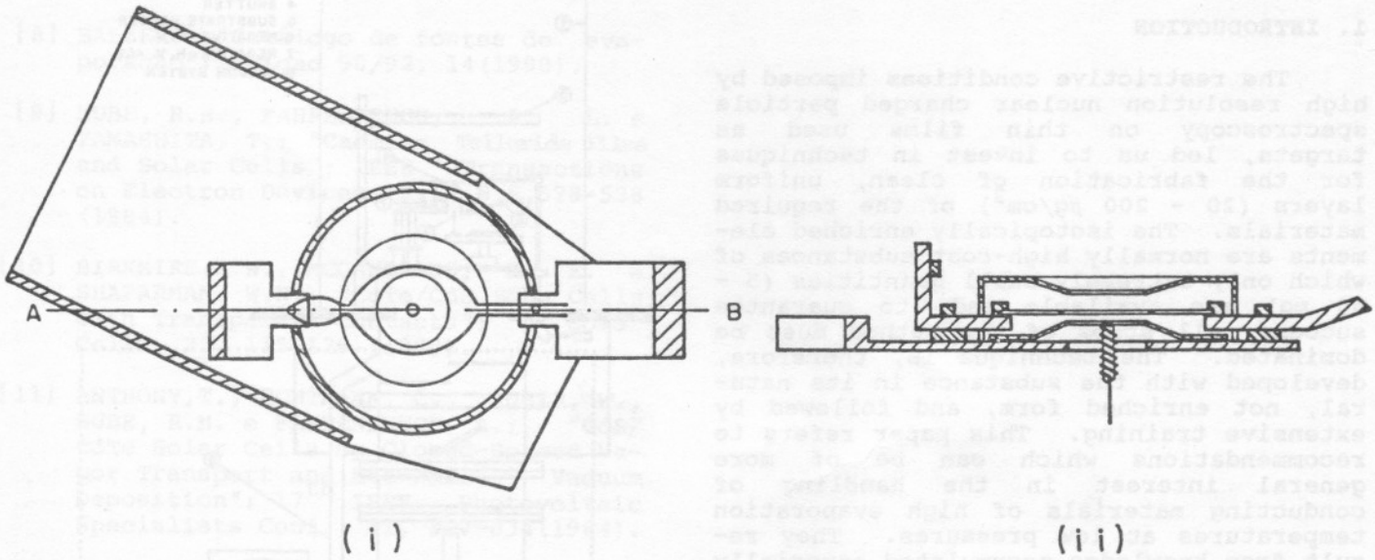

Fig.2. Detailed views of the bombarding system:

(i) horizontal section in the plane of the filament.

(ii) vertical section in the plane a-b indicated in (i).

isotope, the following steps were executed in vacuum. The filament, especially if new, was slowly heated, observing the vacuum, until the collimator appeared dull-red. Several intermediate cool-downs were normally necessary till, at the operating voltage of the filament, the base-vacuum was again obtained. Thorough heating of the rest of the electron bombardment pieces was also promoted by slowly elevating the high voltage, mantaining the crucible at a position higher than usual, and observing the vacuum. When the collimator and the crucible reached temperatures where the base-vacuum was no longer attainable the process was stopped. As a last step, plasma glow cleaning was applied on the whole system, including the crucible, in a nitrogen atmosphere. A complete cool-down is essential before each breaking of the vacuum.

3.2. The geometry of the E.B. system and the control of the focus

It was found that the geometry of the filament - collimator - crucible system is extremely critical in obtaining an adequate focus on the material. The filament should be planar and well shielded by the collimator. The collimator must be clean, without edges, points or cuts due to previous short-circuit problems. The format of the crucible, which serves as anode, is crucial if the electrons are to impinge on the conducting evaporant and not on the crucible, which, made of W or Mo, has evaporation temperatures not much higher. It was found that the crucible must be of comparably small diameter (in the E25016 geometry, less than 3.5 mm, which corresponds to a ratio of 1:5 to the collimator diameter) and have an almost planar top. Since this geometry limits the quantity of material which can be put on the top of the crucible, experiments with different geometries were performed. By weighing the crucible before and after the evaporation and observing its surface critically, it was found that the attempt of deepening the recess on the top resulted in the impossibility of evaporating all of the material, part of it forming an alloy with the crucible material. A cuniform recess gave the worst results. On the other hand if larger-diameter crucibles were employed, it was extremely difficult to maintain the focus on the evaporant and usually some of the crucible material, especially from the edges, also was evaporated.

3.3. The heating-up of the material

Most of the isotopes are delivered in powder form by the fabricant. They, therefore, deserve special care in heating-up in order not to be lost. Starting from a good base-vacuum the temperature in the region of the crucible is slowly elevated by, initially, applying power only to the filament. An increase in pressure indicates that low evaporation temperature substances (absorbed gases mostly) are liberated. Passing too rapidly through these points almost invariably results in "explosions" of gas "bubbles" and loss of material. The power is only increased when the base-vacuum is again reached. The heating-up proceeds by very slowly applying high-voltage, after returning the filament to its work-voltage. Normally at several well-defined points, for each evaporant, a liberation of contaminating material is observed and controlled in the manner described above. The shutter must be closed throughout this procedure. If an appreciable heating of the surroundings is detected by a warming of the pyrex bell jar, the sequence is interrupted and continued only after a complete cool-down.

At about 4-5 W of bombarding power it is possible to "see" the focus on the material, which, if observed through a dark filter, is barely luminous at the points where the electrons impinge. By slowly moving the crucible up and down the best situation is obtained.

With some materials a "flying-off" of the powder particles (which seemed to be charged) along the field lines was observed, at very low power. This was overcome by starting the process with the crucible well below the focus point, letting it remain there in the critical low power range (below 4 W), after which it was returned to the focal position.

3.4. The evaporation

In the trial evaporations with the substances in their natural form it is extremely important to adequately characterize the power input where fusion occurs. After the best focus was obtained, the evaporant was continously monitored through the dark filter. Fusion has occurred if a droplet is formed in place of the little "mountain" of material. At this point the shutter must be removed and it is best to evaporate the material sud-

denly by a rapid elevation of the power up to 160-180 W. The evaporation is characterized by an abrupt increase in pressure. Thirty to sixty seconds is usually sufficient to liberate all of the material. Immediately the shutter should be closed. If the power is elevated too slowly at this point or if the focus is not adequate, the droplet wets the surface of the crucible and a high evaporation temperature alloy may be formed. If the maximum power is maintained too long, the substrates are subject to burns and crucible material may be evaporated. Three to five subsequent evaporations of the material (3-5 mg may be put onto the crucible top) were usually necessary to produce targets with adequate thicknesses.

3.5. Precautions with the substrates

The vapor was condensed on thin carbon films previously evaporated through a similar E.B. or by a carbon-arc method onto glass slides, treated with a releasing agent. Betain was found to produce crystals at the temperatures to which it must be exposed. An extremely thin layer of RBS detergent gave the best results in posterior floating off of the films. If the RBS layer was not sufficiently uniform and thin, bubbles would be formed during the several exposures which exploded the C layer.

The substrate holder had to be positioned 6 - 8 cm above the crucible to minimize burns and in order to guarantee approximately uniform regions large enough for the 1 cm diameter targets. The central region of about 3 cm diameter was, nevertheless, sometimes subject to excessive heating in which case it was impossible to float the film off the slide. The use in this central region of masked C films, already stretched on the target frames, normally produced good spot targets.

If no additional precautions were necessary to avoid oxidation of the targets, it was observed that letting the RBS rehidrate in normal atmosphere during several days eased the floating off, which followed the usual procedure.

4. ACKNOWLEDGMENTS

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