Bull. Inst. Chem. Res., Kyoto Univ., Vol. 47, No. 2, 1969

Vacuum Evaporator with an Electron Gun for the Preparation of Thin Target

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Received March 10, 1969

An apparatus with an electron gun was designed and constructed in order to prepare thin films used as targets in low energy nuclear physics experiments. With this apparatus, self-supporting thin films of 0.1~1 mg/cm^2 thick and 10~30 mm in diameter have been prepared from substances including lithium to bismuth. Targets prepared and coatings and soaks used are tabulated.

I. INTRODUCTION

Thin films of various substances are always required as targets in low energy nuclear physics experiments. The target preparation is an essential step in the experiment. A preparation of self supporting films is a guarantee to simplify experimental procedures and is particularly required in the use of an enriched isotope or of an element with the abundance ratio of 100 percent.

In our laboratory, the film preparation has been performed with the filament heating method,[1] but the application of this method is limited to substances of low evaporation temperature. The electron bombardment method is suitable for the film preparation of various substances. A vacuum furnace with an electron gun can be used as an evaporating source.

The reasons for using a vacuum system for film deposition are summarized as follows:

i) There is an increase of the mean free path of vapour molecule in vacuum.
ii) There is a reduction of the formation of oxides on the evaporating surface of substance.
iii) There is a reduction of contamination in the deposit.

An apparatus easy to handle and suitable for the film preparation was de-

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signed and constructed under the joint-work of one of the authors (Y. U.) and the members of Shimadzu Seisakusho Ltd.. Since 1965, the apparatus has been in operation and self supporting films of various substances have been easily produced by many reseachers. With these films several experiments on the nuclear reaction and the charge exchange of heavy ions have been performed at Kyoto University. This report contains the description of the apparatus and of procedures of the film preparation. A part of the latter was reported in the circulars in Japanese.[2,3]

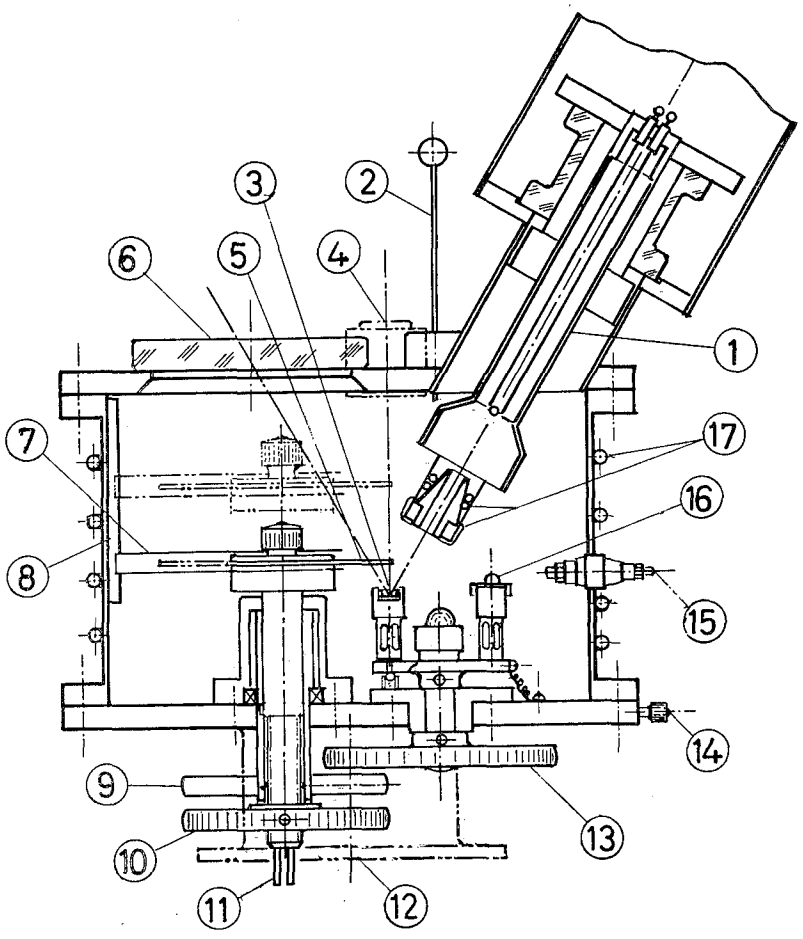

Fig. 1. Diagrammatic view of the evaporation chamber, vacuum seals omitted. (1) Electron gun, (2) Hook, (3) Melting pot, (4) Box for a liquid-nitrogen vessel cooling a substrate holder (not shown in the figure), (5) Substrate holder, (6) Window, (7) Shielding box of the substrate table, (8) Guide rail (9) Handle, turning, (10) Handle, up and down, (11) Cooling pipe, (12) Exhaust tube, (13) Handle, turning, (14) Terminal to gronud, (15) Terminal, up to 70 A, (16) Cap of the pot, (17) Cooling pipes.

II. APPARATUS

In designing the apparatus, the following points were especially taken into account:

i) An electron gun is mounted obliquely to a chamber so that the large space is left for the set of tools such as substrate holders and hooks.

ii) Heating of substances is performed in two steps. At the stage to release absorbed gases from the substance, the melting pot is bombarded with electrons and the substance in the pot is heated through the wall of the melting pot and at the stage of evaporation, the substance is directly bombarded with electrons. Therefore, the melting pot is required to move on either side of the fixed focus point of energetic electrons.

iii) The aperture of windows should be large to put tools easily in and out of the chamber.

iv) Circular handles to move holders should be of large diameter in order to free operators from fool operation.

Figure 1 shows a diagrammatic view of the evaporation chamber of 37 cm in inner diameter and 24 cm in height. In the figure, omitted are vacuum seals and overlapping parts.

An electron gun (1) is attached obliquely to a lid of the chamber and electrons are incident at 30° on a melting pot (3). A substrate holder is set opposit to the electron gun. The substrate holder is mounted on a circular table (5) which is rotated with a large circular handle (9) from the outside of the chamber. The distance between the substrate holder and the melting pot can be varied from 3 to 9 cm with a large circular handle (10). Three pots are placed on stands mounted on a table, which is rotated with a large circular handle (13). The substrate table is shielded from the vapour with a box (7) when the substance is heated to release absorbed gases. Unused pots are also shielded with caps (16), which can be removed with hooks.

In addition to the elements described above, constituent parts fitted to the

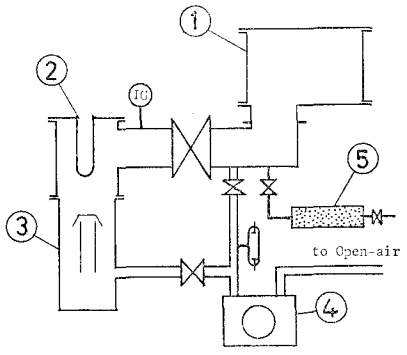

Fig. 2. Schematic diagram of the vacuum system.
(1) Evaporation chamber, (2) Liquid-nitrogen trap, (3) 700 l/sec oil diffusion pump, (4) 300 l/min mechanical pump, (5) Silica-gel desiccator.

chamber are: a circular window (6) of 15.3 cm in diameter, two hooks (2) with sphere joints for tilting and sliding, a substrate holder cooled with liquid nitrogen, a vacuum gauge box and a lamp case box on the lid, the holders for the melting pot, the substrate holder and a vacuum exhaust tube (12) on the bottom, and finally, a rectangular window 20 cm×9.8 cm in inner size, a vacuum gauge box and three electric terminals (15) capable of carring a current up to 70 A on the side wall.

Figure 2 shows a schematic diagram of the pumping system. An oil diffusion pump with 4 stage nozzles and fractionating parts is joined through a liquid nitrogen trap to the tube attached to the bottom of the chamber. This exhaust tube is of 16 cm in inner diameter.

The volume of the vacuum chamber is about 30 liters. The inner surface of the chamber is of 3×10^3 cm^2 in area and the surface of the tools contained in the chamber has nearly the same area. The puming down time of this vacuum system is about 30 minutes from 1 atmosphere to $3\sim4\times10^{-6}$ torr without the use of the liquid nitrogen trap. The pressure below 1×10^{-5} torr can be obtained in the deposition period.

The electron gun is of a single self-focusing type used in the industrial x-rays tube. The filament for electron source is of a coil type of 19 turns, 0.9 mm in inner diameter and 10 mm in length, made of the wolfram wire of 0.2 mm in diameter. The cathode electrode has approximately a concave shape of 34 mm in radius and 46 mm in curvature radius. The distance from the filament to the

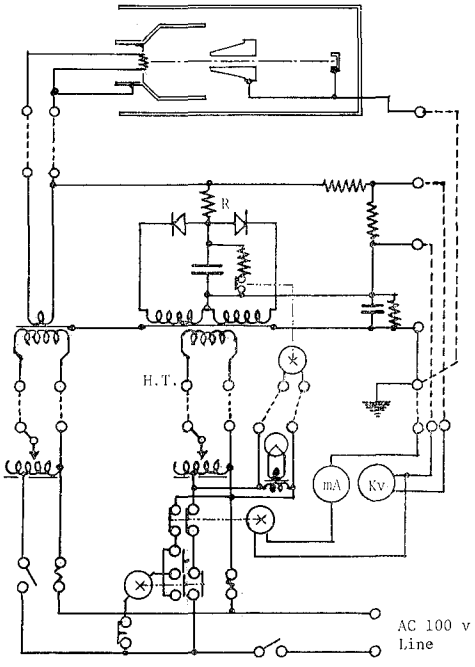

Fig. 3. Circuit diagram of the electron gun.
Filament power: 15V×6A, High-tension
Power: 50kV×50mA, R; 60kΩ, 200W.

Table 1. List of Prepared Targets.

1 Substance Z	2 Self-support or Not	3 Thickness [mg/cm²]	4 Size [mmφ]	5 Backing	6 Substrate	7 Coating	8 Soak [time]	9 Note
3 Li	Self	0.1～0.3～0.5	20～30		Glass		Oil [Kerosene] [10 min]	Fill up dry A-gas, strip off with a razor
LiF	Self							
4 Be	Self	0.3, 0.05～1			Glass	Nitro-Cellulose	Water→Acetone	Contami : C & O [5%]
	Self				Glass	NaOH	Water	
5 B	Self	0.05～1	8～12		Glass		Water	
					Glass	Tee-pole	Water	
6 C	Self	～0.01	～10		Glass	KOH [a]	Water	[a] better than [b] and [c]
	Self	～0.01	～10		Glass	Tee-pole [b]	Water	
					Glass	Polystyrene (c)	Benzen	
9 F (LiF)	Self or Not	0.01～0.5	～10		Ni or Al		Water	
11 Na	Self	1	10～15		Glass		Oil [Kerosene] [10 min]	Fill up dry A-gas, strip off with a razor
12 Mg	Self	0.3	20～30		Glass	Nitro-Cellulose	Water→Acetone [5 min]	Contami : C & O [5%]
					Glass	Formvar		
13 Al	Self	0.1	15		Glass			
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
					Glass	Formvar		
14 Si	Self	0.5	～10		10μ-Al		NaOH [20min]	Contami : Na & Al [small], O [5%]
					Glass	Formvar		
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	In high vacuum

16 S	Not			Au etc.				Sandwich between metal foils
20 Ca	Self	1	20～30		Glass		Oil [Kerosene]	Contami : O [small], strip off with a razor
21 Sc	Not	5		Pt				
22 Ti	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	In high vacuum
23 V	Not	5		Pt				
24 Cr	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water [hot]	In high vacuum
25 Mn	Not	0.1	——	Formvar				
26 Fe	Not			Formvar				
29 Cu	Self	0.1	15～30		Glass	Formvar		
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
30 Zn	Self	1	——		Glass	Formvar		
32 Ge	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	Stripping difficult
33 As	Not			Formvar				
34 Se	Self	1	——		Glass	Formvar		
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
40 Zr	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
47 Ag	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
48 Cd	Self	1	15		Glass	Formvar		
50 Sn	Self	1			Glass	Formvar		
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	Stripping difficult
79 Au	Self	0.1	20～30		Glass			
	Self	0.01～0.1	～10		Glass	NaCl or KCl	Water	
82 Pb	Self	1	30		Glass	Formvar		
					Glass	Nitro-Cellulose	Acetone	
83 Bi	Self	1	30		Glass	Formvar		

anode electrode is 1.2 times as long as the curvature radius of concave. A central hole of the anode electrode is of 11 mm in diameter. An image of electron beams focused at the pot was found to have an oval shape of 1.6 mm×3.2mm.

A melting pot is made of wolfram. The charging volume of the pot is of 0.2 ml, 8 mm in diameter and 4 mm in deepness. The thickness of the wall is required to be 3 mm so that the focusing electron bombards the pot without bombarding the sample. Two of the three pots can be used for the deposition of coating substances.

The side wall of the chamber and the anode electrode are cooled with water. The substrate table conducts the heat to its shaft cooled with water.

Figure 3 shows an electrical circuit diagram of the electron gun. The power supply for the electron gun and for the filament are of 50 kV×50 mA and 15 V× 6 A, respectively.

III. PERFORMANCE

III-1. X-rays radiation

X-rays are produced by the decceleration of energetic electrons when they strike a target. In the initial test, an intense radiation of 245 mr per hour was observed at the outside of the glass-plate window 12.5 mm in thickness with a survey meter (ionization chamber type). Using a glass plate 25 mm in thickness, the radiation dose was observed to be nearly zero in the full scale of 3 mr per hour under operating condition of 10 kV×10 mA.

III-2. Target preparation

Targets prepared with this apparatus are listed up in Table 1. In the table, the seventh and eighth columns show the condition to prepare the self-supporting film. The substance is deposited on the substrate coated with the coating (7 th col.) and the film is stripped off from the substrate in the soak (8 th col.). Combinations of the coating and the soak are summarized especially in Table 2.

The soak has the following characteristics:

i) The coating is soluble in the soak.
ii) The substance is not soluble in the soak.
iii) Water is often used to float the thin film on the water surface with its strong surface tension.

In the case of Be, for example, nitrocellulose is used as a coating to utilize the characteristic iii) for stripping the film from the substrate. Nitrocellulose itself is solved in acetone.

III-3. Discussion of the vapour source

In order to give a simple outlook of the evaporation system in operation, characteristic features of the vapour source are discussed in the following.

The rate of evaporation in vacuum is given by[4]

$$5.85\times10^{-2}\times k\,P\sqrt{\frac{M}{T}}\,\mathrm{g\cdot cm^{-2}\cdot sec^{-1}},$$

where P (torr) is the vapour pressure at T(°K), M a mol of the substance being

Table 2. Combination of the Coating and the Soak to Produce Self-Supporting Films.

Sub-strate	Strip off		Dissolve	Note
	Coating	Soak	Solvent	
Glass	Non	Water,	——	
	Non	Oil (mechanically)		Oil : free from air [O_2 and H_2O vapour]
	Nitro-Cellulose	Acetone	——	Nitro-Cellulose 5~10 % [Acetone solution], coat and dry
	Nitro-Cellulose	Water	Acetone	
	Poly-styrene	Benzen	——	
	Tee-pole	Water	——	Tee-pole 5~10 % water solution], coat and dry
	Tee-pole	Ethyl alcohol		Ethyl alcohol 30 % [water solution]
	Cleanser	Water	——	Cleanser 5~10 % [water solution], coat and dry
	NaCl	Water	——	Evaporate crystalloid NaCl
	KCl	Water	——	Evaporate crystalloid KCl
	HaOH	Water	——	NaOH 10 % [water solution], coat and dry
	KOH	Water	——	KOH 10 % [water solution], coat and dry
	Formvar	Water	Ethylene dichlorid [$C_2H_4Cl_2$]	Formvar 5~10 % [$C_2H_2Cl_2$ solution], coat and dry
	NaOH-Formvar	Water	Ethylene dichlorid [$C_2H_4Cl_2$]	
Al	Non		KOH	KOH 10% [water solution, hot]

Table 3. Typical Operating Condition of the Electron Gun.

Substance	High tension [kV]	Current [mA]	H.T.× Curr. [W]	Melting temp.[6] [°K]	Temp. at 0.1 torr[6] [°K]	$\sqrt{M/T}$ [at 0.1 torr]
Li (natural)	10	3~4	30~40	454	896	8.8×10^{-2}
LiF	7	6	42	[1143]		
$B^{10,11}$	15	16~20	240~300	2300	2650	6.3×10^{-2}
Al	9	10	90	932	1620	1.3×10^{-1}
Si	10	13	130	1688	1990	1.2×10^{-1}
V	13	10	130	2130	2310	1.5×10^{-1}
Ni	10	13	130	1725	1940	1.8×10^{-1}

evaporated, and k the condensation coefficient which has been found to be equal to unity for most metals.[5] Typical operating conditions of the electron gun system are shown in Table 3.

Let's consider the evaporation of natural Li metal and assume that the metal is evaporated at the temperature corresponding to the vapour pressure of 1×10^{-1} torr. The temperature is 900 °K for Li metal. Then the rate of evaporation is 5.2×10^{-4} g·cm^{-2}·sec^{-1}. The calculated thickness for the film deposited onto the

unit area, just above and 8 cm apart from the surface source (0.5 cm^2 in area), is 0.16 $mg \cdot cm^{-2}$ in 2 minutes, which should be compared with the obtained thickness of 0.2 $mg \cdot cm^{-2}$.

The power needed for the evaporation of Li metal was estimated as follows:

i) Conduction loss is estimated to be very small because of the small contact area between the melting pot and its stand.

ii) Radiation loss is given by the Stefan-Boltzmann formula,
$E_R = 5.8 \times 10^{-12} \cdot T^4$ $W \cdot cm^{-2}$,
where T(°K) is the temperature of the substance. Since the surface area of the melting pot is approximately 5.7 cm^2, the total radiation loss for Li metal at 900°K is estimated to be 22 W.

iii) The heat of vaporization of Li metal at 1590°K is 35.4 $kcal \cdot g\text{-}atom^{-1}$ at the boiling point (~1600°K). Then, the energy loss corresponding to the vaporization rate mentioned above is estimated to be 2.0 W.

The total power needed is thus approximately 24 W, which should be compared with the supplied input power of 30~40 W.

Summing up the above mentioned features, i) the heat energy is almost lost by radiation and ii) for the substances of low melting point such as Li metal, the vapour pressure in the deposition period is about 1×10^{-1} torr.

For more convenient operation of the apparatus, the following improvements are hoped:

i) The image size can be varied with an electrostatic lens in order to perform easily the two steps of heating.

ii) The substrate holder is sufficiently cooled with water.

ACKNOWLEDGMENTS

The authors would like to express sincere thanks to Prof. Takuji Yanabu, Prof. Sukeaki Yamashita, Prof. Isao Kumabe, Dr. Kiyohiko Takimoto, Prof. Masakatsu Sakisaka and Prof. Fumio Fukuzawa for their advices and encouragements.

They are indebted to Dr. Tetsumi Tanabe, Dr. Kazuhiko Hosono and Dr. Yasuhiko Okuma for their discussions and suggestions to prepare the targets.

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