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VAPOR DEPOSITED FILM DISTRIBUTION ON
CURVED SUBSTRATE SURFACES

by

R. L. Beno

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

THE DOW CHEMICAL COMPANY
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ABSTRACT

A comparison between calculated and experimental film thickness distribution on curved substrate surfaces is described. Using copper as evaporant, the emission characteristics of a tantalum crucible and a surface emitting pan source are compared. Two simultaneously emitting tantalum crucibles, situated beneath the exterior of a hemispheric substrate surface, did not upset distributions as determined by Knudsen's cosine law.

SUMMARY

1. An upward evaporating crucible can be considered as a surface emitting source for predicting distribution coefficients.
2. Experimental film thickness distributions obtained on a 4-1/2-inch radius curvature, from two surface emitting sources, compares favorably with the calculated distributions.

INTRODUCTION

In vacuum vapor deposition of metallic films, thickness distributions on plane or curved substrate surfaces can be precalculated using Knudsen's cosine law. This law for evaporation involves the cosine of the emission angle for any direction under investigation. To obtain a specified film distribution, one has to consider the shape of the receiving surface and the arrangement of the vapor source or sources relative to the surface. For Knudsen's cosine law to be applicable, Holland⁽¹⁾ lists three conditions which must be assumed. These are:

1. The evaporation is carried out at sufficiently low gas pressure for the scattering of vapor molecules by collisions with gas molecules to be negligible.
2. The vapor intensity is so low that the effect of collisions between vapor molecules in the vicinity of the source can be neglected.
3. That every vapor molecule striking the receiving surface condenses on first impact.

Numerous investigators have shown the validity of the cosine law on plane substrates. Behrndt⁽²⁾ used the principle to construct a shutter which would alter the

distribution from a source with nonuniform thickness characteristics. Strong⁽³⁾ used the technique to deposit a film of varying thickness on a spherical surface. A comparison was made in this laboratory of point source distribution on plane substrates at incidence angles of 90°, 75°, and 60°. No references have been found, however, which deal with uniform coating of hemispheric rotating substrates from two or more surface sources operated simultaneously. It seemed worthwhile to investigate this relationship so that one might predict in advance what uniformity could be expected from any specific coating arrangement.

The concept of a surface source differs from that of a point source in the degree of direction of the vapor stream. A point source is considered a small sphere, evaporating material at an equal rate, in all directions. A surface source is a small area from which material is evaporated at a specific rate from one side only.

The purpose of this work was to determine if the cosine law calculations would hold for dual sources evaporating material simultaneously to a curved substrate surface. This is accomplished by (a) showing that a crucible can be considered a surface emitting source, (b) calculating the distribution pattern for a single crucible evaporating material to a curved substrate surface, (c) calculating distribution patterns for dual crucibles evaporating material to a hemispheric surface, and (d) comparison of the experimental and calculated distributions.

EXPERIMENTAL

An Allen-Jones BA65* tantalum crucible, resistance heated, was used singly and dually for the evaluation experiments. The crucibles were mounted through an auxiliary base plate to achieve upward evaporation. Copper was the evaporant material.

Initially, a comparison was made between the crucible vapor pattern and the vapor pattern of a dimpled pan source to determine if the crucible could be considered a surface emitting source. The substrate for this evaluation was a flat strip of aluminum foil 4 inches from the source.

A curved substrate surface simulating a hemisphere of 4-1/2-inch radius was used for the remaining evaluations. A strip of aluminum foil was attached to this surface to enable measurement of the film thickness distribution. Following the evaporations, the aluminum strip was removed, sectioned into 1/2-inch squares and weighed. The film was then removed and the piece

(1) Holland, L., *Vacuum Deposition of Thin Films*, New York, John Wiley & Sons, 1956, pp 142-3.

(2) Behrndt, K. H., "Films of Uniform Thickness Obtained From a Point Source," 1962 *Transactions of the Ninth National Vacuum Symposium of the American Vacuum Society*, G. H. Bancroft, ed., New York, MacMillan Company, 1962, pp 111-5.

(3) Strong, J., *Procedures in Experimental Physics*, New York, Prentice-Hall, Inc., 1938, pp 174-5.

* Allen-Jones Electronics Corp. 17171 South Western Ave., Gardena, Calif.

tared. The thickness was calculated using $-\frac{[\text{surface density (g/cm}^2\text{)}]}{[\text{specific density of bulk copper (g/cm}^3\text{)}]}$.

The mounting of the substrate relative to the sources is shown in Figure 1 through Figure 4.

For the dual evaporations, it was essential that both crucibles be operating at the same temperature. Since separate crucible-heater power supplies were used, and because of possible resistance variations between the two crucibles, a check was made by infrared pyrometer for correct current-temperature settings. Crucible temperature was 1300°C.

EVALUATION I - Distribution Pattern Comparison Between a Crucible and Flat Pan Source.

Because of possible directional effects, it was necessary to determine whether or not a crucible could be considered a surface emitting source. To determine this, it was compared to a source which closely simulated a theoretical surface source. The flat pan source consisted of a flat strip of tantalum with a 1/2-inch diameter by 1/16-inch deep flat dimple to contain the evaporant material.

Figure 1 shows the arrangement for comparing the two sources. After completing the crucible run, the pan source

was substituted in the same location as the crucible and loaded with an identical weight of evaporant material. The thickness of the condensed copper films, from both evaporations, was measured at 1/2-inch intervals, from the center out 4 inches on each side. Table I gives the film thickness values for both determinations and Figure 2 is a plot showing the similarity of the two patterns.

EVALUATION II - Film Distribution Patterns for a Crucible Evaporating Material to the Exterior of a Curved Substrate of 4-1/2-inch Radius.

Two separate pattern determinations were made with the crucible in different positions beneath the substrate. For these evaluations the substrate was positioned above the crucible source with the equator inclined to a 45 degree angle. The lower quadrant was then the condensing surface. Figure 3 and Figure 4 show the positions of the crucibles for the separate pattern evaluations. Table II shows the film thickness values obtained from these determinations. Figure 5 is a comparative plot of the crucible patterns obtained from the apparatus as depicted in Figure 3 and Figure 1 showing the variation between a plane and curved substrate. The variation begins to be evident at 2 inches from center, where the thickness begins to decrease more rapidly for the curved substrate surface.

Figure 1. CRUCIBLE TO PLANE SUBSTRATE

Figure 2. CRUCIBLE vs FLAT PAN SOURCE

Figure 3. CRUCIBLE TO CURVED SUBSTRATE

Figure 4. CRUCIBLE TO CURVED SUBSTRATE

TABLE I

FILM THICKNESS VALUES ON A PLANE SUBSTRATE
FROM A CRUCIBLE AND FLAT PAN SOURCE

Substrate Position	Film Thickness From Crucible Evaporation (Å)	Film Thickness From Pan Evaporation (Å)
Center	14,500	15,600
1/2 inch from center	13,900	14,300
1 inch from center	10,600	11,900
1-1/2 inches from center	7,600	8,600
2 inches from center	5,200	5,900
2-1/2 inches from center	3,800	4,600
3 inches from center	2,600	3,100
3-1/2 inches from center	1,900	2,200
4 inches from center	1,200	1,600

Figure 6 is a plot of the film distribution from the crucible in Figure 4 position, or directly beneath the equator. A marked difference in the pattern is observed from that of the first crucible position (Figure 3). Film thickness increases from Point A and is greatest where the plane of

the substrate is perpendicular to the direction of travel of the vapor. From there, the thickness decreases at a slightly greater rate toward the pole because of the greater inclined angle of the substrate.

TABLE II

FILM THICKNESS VALUES FROM A CRUCIBLE IN TWO POSITIONS
BENEATH A CURVED SUBSTRATE SURFACE

Figure 3 Crucible Position

Substrate Position	Film Thickness (Å)
Center (c)	14,300
1/2 inch from center	13,100
1 inch from center	9,600
1-1/2 inches from center	6,800
2 inches from center	4,900
2-1/2 inches from center	3,200
3 inches from center	2,400
3-1/2 inches from center	800
4 inches from center	200

Figure 4 Crucible Position

Substrate Position	Film Thickness (Å)
Equator (A)	9,000
1/2 inch from A	11,400
1 inch from A	12,500
1-1/2 inches from A	11,900
2 inches from A	8,700
2-1/2 inches from A	5,500
3 inches from A	3,100
3-1/2 inches from A	1,700
4 inches from A	1,000
4-1/2 inches from A	700
5 inches from A	300

Figure 5. DISTRIBUTION COMPARISON OF CURVED SUBSTRATE vs FLAT SUBSTRATE

Figure 6. DISTRIBUTION FROM CRUCIBLE AT POSITION A

EVALUATION III – Comparison of Calculated and Experimental Film Thickness Distribution Resulting from Two Crucibles Emitting Metal Vapor Simultaneously.

For purposes of precalculating the film thickness, using the cosine law, for dual crucibles evaporating material to the

exterior of a hemisphere of 4-1/2-inch radius, consider the diagram in Figure 7. Positioning one crucible directly beneath the equator and the other crucible on a vertical line beneath the pole provides the best arrangement for greatest uniformity of the resulting film.

Figure 7. TWO CRUCIBLE SOURCES POSITIONED BENEATH POLE AND EQUATOR OF HEMISHELL

Consider, first, the hemisphere as being stationary. Assume, also, that copper with a density (ρ) of 8.92 g/cm³ to be the evaporant material and the average desired thickness of the film to be 50,000 angstroms (0.2 mil). To ascertain the mass of material (m) required to produce this average thickness, we must assume some value greater than 50,000 angstroms for point t_2 (Figure 7). From the information obtained in Evaluation II, the thickness at t_1 will be approximately 80 percent of t_2 , so we can calculate the mass assuming a 60,000 angstrom thickness at t_2 .

$$m = \frac{\bar{A} \cdot \pi \cdot \rho \cdot (r)^2}{\cos \phi \cdot \cos \theta \cdot 10^8} = 1.6 \text{ grams of copper} \quad (\text{Eqn. 1})$$

Lambert's law for radiation expresses the cosine law of illumination and the inverse square law.

$$E = \frac{I \cos \theta}{r^2} \quad (\text{Eqn. 2})$$

I = photometric intensity

θ = angle at which the illuminated surface subtends at the point source of light

r^2 = the square of the distance from the point source of light to a point on the illuminated surface.

From this Holland⁽¹⁾ derives the basic equation for a surface evaporating source.

$$t = \frac{m}{\pi \rho} \cdot \frac{\cos \phi \cdot \cos \theta}{r^2} \quad (\text{Eqn. 3})$$

ϕ = Angle at which the receiving surface deviates from a normal

θ = Angle at which the receiving surface is inclined to the direction of the vapor stream

r^2 = The square of the distance from the emitting surface to a point on the receiving surface.

Since crucibles S_1 and S_2 are equidistant and at the same angle to point t_3 , we can assume the same mass of copper in each crucible for calculating any point on the curvature, which lies within line of sight of the two sources.

$$t_1 \text{ and } t_5 = \frac{m}{\pi \rho} \cdot \frac{\cos \theta_2}{(h)^2} \cdot 10^8 = 34,000 \text{ \AA} \quad (\text{Eqn. 4})$$

$$t_2 \text{ and } t_4 = \frac{m}{\pi \rho} \cdot \frac{\cos \phi \cdot \cos \theta}{(r)^2} \cdot 10^8 = 60,200 \text{ \AA} \quad (\text{Eqn. 5})$$

Since point t_3 will be receiving equal vapor from crucibles S_1 and S_2

$$t_3 = 2 \left[\frac{m}{\pi \rho} \cdot \frac{\cos^2 \theta_1}{(r_1)^2} \cdot 10^8 \right] = 43,200 \text{ \AA} \quad (\text{Eqn. 6})$$

Point t_6 will receive vapor from crucible S_2 only

$$t_6 = \frac{m}{\pi \rho} \cdot \frac{\cos \phi_2 \cdot \cos \theta_3}{(r_2)^2} \cdot 10^8 = 5700 \text{ \AA} \quad (\text{Eqn. 7})$$

Some vapor will arrive at t_2 from crucible S_2 and likewise at t_4 from crucible S_1 . This was calculated to be less than 2000 angstroms, so was disregarded. The symbols used in the above equations are shown in Table III with their corresponding values. Theoretical values were obtained for other points at each 1/2 inch along the substrate and are given in Table IV.

TABLE III

VALUES FOR SYMBOLS USED IN CALCULATIONS

Symbols	Length (cm)	Symbols	Cosine	Angle
h	10.9	θ (theta)	.9998	1°
r	9.3	θ_1	.6820	47°
r_1	11.08	θ_2	.7071	45°
r_2	14.9	θ_3	.2250	77°
		ϕ (Phi)	.9135	24°
		ϕ_1	.6820	47°
		ϕ_2	.9868	9° 20'

(1) Holland, L., *loc. cit.*, p 1.

TABLE IV

COMPARISON OF EXPERIMENTAL WITH
CALCULATED FILM THICKNESS VALUES

Substrate Position (Figure 7)	Experimental Thickness (Å)	Calculated Thickness (Å)
A (t ₁)	35,100	34,000
1/2 inch from A	47,500	46,200
1 inch from A	59,900	57,000
1-1/2 inches from A	59,900	61,700
t ₂		60,200
2 inches from A	49,600	57,200
2-1/2 inches from A	45,500	46,500
3 inches from A	45,600	44,800
3-1/2 inches from A	49,100	43,200
t ₃		44,800
4 inches from A	50,700	44,800
4-1/2 inches from A	58,200	46,500
5 inches from A	59,900	57,200
t ₄		60,200
5-1/2 inches from A	58,700	61,700
6 inches from A	48,300	57,000
6-1/2 inches from A	34,100	46,200
7 inches from A (t ₅)	28,800	34,000
7-1/2 inches from A	15,500	

The crucible arrangement shown in Figure 7 was simulated experimentally for verification of this calculated pattern. Following the simultaneous evaporation, thickness measurements were made every 1/2 inch, starting at t₁ and continuing through t₅. Table IV shows the values obtained.

Figure 8 is a plot of these values showing the similarity of the experimental with the calculated thicknesses. Precision of the experimental values from t₁ through t₅ was 50,000 angstroms $\pm$ 17,000 angstroms.

DISCUSSION

By rotating the substrate, about the pole axis, the entire exterior surface of the hemisphere could be coated. The amounts of evaporant material in the crucibles would be increased proportionately to provide for the increased surface area of the substrate. Some approximation could be made of this increase in mass of copper required to provide a 50,000 angstrom film thickness at point t₅, the only stationary point during rotation.

$$m = \frac{50,000 \times 1.6}{28,800} = 2.78 \text{ grams copper (crucible } S_2) \quad (\text{Eqn. 8})$$

The points t₆ and t₄ would be interchanging during rotation, so the resulting film thickness for points around this circumference would be the average of the t₆ and t₄ thicknesses

Figure 8. COMPARISON OF EXPERIMENTAL AND
CALCULATED FILM DISTRIBUTION

calculated while the hemisphere is stationary. A similar deduction could be used for point t₃ which interchanges with a point opposite to it on its circumference. This opposite point, however, receives no vapor when the hemisphere is stationary. The resulting thickness at point t₃, from crucible S₂, would be one-half the calculated thickness at point t₃ when stationary.

To find the mass of copper required in crucible S₁, different reasoning is applied. There is no stationary point, within the sphere of radiation from this crucible, when the hemisphere is rotating. Calculations are based on the circumference on which t₂ is a point, as shown in Figure 9. This circumference is parallel to the equator. For purposes of calculation, we could assume three additional crucible sources, at each other quadrant of the hemisphere, situated in the same relation as crucible S₁. Figure 10 shows this configuration. If each of these crucibles contained the same amount of copper (1.6 g), and all four simultaneously evaporated to exhaustion, the resulting film thickness at any point on this circumference would be the average of the thicknesses obtained by calculating four points, 22.5 degrees apart, of one quadrant. In Figure 10, these points are t₂, t₇, t₈, and t₉. This average could be assumed to be representative due to rotation of the substrate.

The calculations would be made for these points with the hemisphere stationary in the same manner as described on page 5.

The average thickness would be less than 60,000 angstroms, and was calculated to be 35,000 angstroms. Since t₂ is the

Figure 9. POSITION OF t_2 CIRCUMFERENCE IN RELATION TO THE EQUATOR OF THE HEMISPHERE

closest point to crucible S_1 , and receives the greatest thickness, it would be necessary to increase the mass of copper to bring this thickness up to 60,000 angstroms, and multiply this amount by four to provide corresponding film thickness over that portion of the hemisphere covered by crucible S_1 .

The actual mass was calculated as follows:

$$m = 4 \frac{60,000 \times 1.6}{35,000} = 10.97 \text{ grams of copper}$$

The thickness range over the entire exterior surface of the hemisphere, considering both sources, would then be theoretically 35,000 angstroms to 60,000 angstroms.

Figure 10. HYPOTHETICAL ARRANGEMENT OF FOUR CRUCIBLES AROUND THE CIRCUMFERENCE ON WHICH t_2 IS A POINT

Combining the masses of copper from both crucibles results in a total of 13.8 grams of copper. A theoretical uniform coating of 50,000 angstroms would weigh 3.7 grams, so the process would be approximately 27 percent efficient in terms of material used.

This hypothesis for the rotating substrate was not proven experimentally as was the uniformity for the stationary curved substrate.

ADDITIONAL REFERENCE

Born, M., and Wolf, E., *Principles of Optics*, 2nd rev. ed., Oxford, New York, Pergamon Press, 1964, pp 181-3.

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