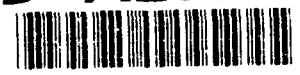

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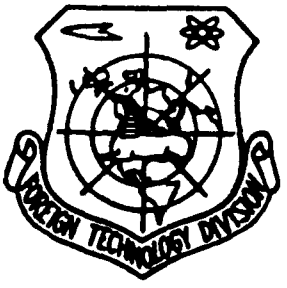
FOREIGN TECHNOLOGY DIVISION

A METHOD OF CALCULATING SUBLIMATION RATE FOR THERMIONIC CATHODES

by

Guo Wenxiang



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A METHOD OF CALCULATING SUBLIMATION RATE FOR THERMIONIC CATHODES

Guo Wenxiang, Institute of Electronics, Chinese Academy of Sciences

[Abstract] An improved method of measuring cathode sublimation rates using a crystal oscillator is proposed; in the method, the distance between the crystal (substrate) and cathode (source) is smaller than is usual. Thus, more sublimation products from the cathode (source) can be received by the crystal and consequently the method can serve in measuring lower sublimation rates. Formulas for calculating the sublimation rate are derived and their precision is stated. In the paper, a method of preventing the resonant frequency change caused by temperature changes is also given. The experimental results show that 1Hz of shift in the resonant frequency can be detected when the oscillation frequency is 5MHz.

I. Introduction

By using the crystal oscillator method in measuring sublimation rates of cathodes, generally the cathode is regarded a point sublimation source [1,2]; this is possibly because of facilitating the calculation of the sublimation rate. In other reports, such as in reference [3], the cathode is considered as a flat-element sublimation source; the sublimate receiver is considered as a flat element at a location along the normal direction of the cathode surface.

The article adopts an effective measure for overcoming the effect of measuring the sublimation rate by the oscillation frequency drift (crystal resonant frequency drift) of the crystal because of temperature change. This not only enhances the precision in measuring the oscillation frequency change (crystal resonant frequency change) due to the sublimate; in addition, it allows a closer distance between the crystal and cathode. That is, in a situation of not changing the area of the crystal receiving the sublimate, the quantity of received sublimate per unit time is increased. In this article the cathode is considered as a flat-element sublimation source. Under the conditions of greater area ratio in receiving the sublimate, equations and inaccuracies in calculating the cathode sublimation rate are derived. In addition, the actual application situations are introduced.

II. Premise Conditions of Calculation

It is assumed that an equilibrium state is received between the molecules just sublimated from the cathode, and the sublimated molecules in the outside; at that instant, the sublimation rate is equal to the condensation rate. If the sublimated molecules are not returned, then the sublimation rate (condensation rate) can be measured [4]. Numerically, this is the collision rate of the sublimated molecules against the cathode in the equilibrium state. Computing the collision rate of the flat element by gas molecules in the equilibrium state is the general method of calculating the sublimation rate of flat-element sublimation sources.

In the following, particular situations of cathode sublimation are analyzed. Considering that the upper limit (the maximum rate) of the cathode sublimation rate is smaller than 10^{16}atoms/cm^2/s (if barium atoms are sublimated, this upper limit of cathode rate corresponds to a value smaller than

$2.3 \cdot 10^{-6} g/cm^2/s$), then the corresponding vapor pressure of the cathode sublimate is lower than 10^{-4}torr [5]. If the effective diameter of the sublimated molecules is $5 \cdot 10^{-8}$cm, and the temperature is higher than 1000K, then the mean free path length is greater than 90cm. If the cathode size is the conventional cathode to be measured, and the cathode is in a vacuum, then if the cathode sublimation is considered as molecules in the equilibrium escaping into the vacuum [6] through small holes from the vessel, then because of the condition that the mean free path length is considerably greater than the cathode dimension, the equilibrium state of molecules in the vessel can be considered such that there is no effect of molecules escaping (notwithstanding that the total number of molecules in the vessel is slowly decreasing) and the molecules escaping in the form of effusion. Therefore, the calculation of the sublimation rate can be considered as the calculation of collision number per unit area of the cathode surface by molecules per unit time at the corresponding vapor pressure.

III. Calculation of Cathode Sublimation Rate

Assume that the area receiving the sublimate is a circle with diameter 2a, and that the sublimation area of the cathode is the area element ds, and two circles are perpendicular to the connecting line oo′ of the circles′ center (Fig. 1). Then the collision number (refer to Fig. 1) to ds per unit time for molecules in the three-dimensional angle (the three-dimensional angle of a cone with vertex angle of $2\theta_0$) of the corresponding circle area receiving the sublimate.

From the theory of molecular motion, the number of molecules colliding at ds per unit time for a three-dimensional angle element $d\omega$ with angle θ formed with oo′ is

$$\frac{d\omega}{4\pi} n\bar{v} \cos\theta \, ds, \tag{1}$$

In the equation, $\bar{v}$ is the average speed of molecules and n is the

molecule number per unit volume. By establishing a spherical surface coordinates with point o as the origin, then $d\omega=\sin\theta d\theta d\varphi$. Then the number of molecules per unit time colliding at ds of the origin o of the three-dimensional angle of the corresponding cone with vertex angle $2\theta_0$ is;

$$N_0 = \frac{n\bar{v}}{4\pi} ds \int_0^{2\pi} d\varphi \int_0^{\theta_0} \sin\theta \cos\theta d\theta = \frac{n\bar{v}}{4} \sin^2\theta_0 ds, \qquad (2)$$

In the equation, $n\bar{v}/4$ is the overall molecules in the direction from the upper side of ds, colliding at a unit area ds per unit time. This can also be expressed as $\frac{p}{\sqrt{2\pi mkT}}$ (p is the vapor pressure, k is the Boltzmann constant, m is molecular weight, and T is absolute temperature). Numerically, $n\bar{v}/4$ is equal to the sublimation rate at ds. Then if N_0 is known, the sublimation rate can also be known; however, N_0 can be measured experimentally.

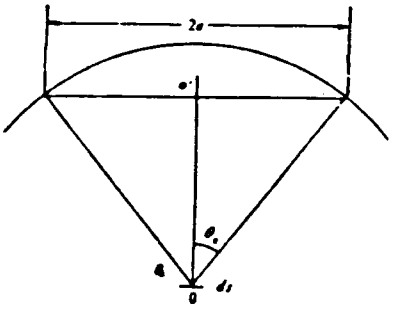

Fig. 1

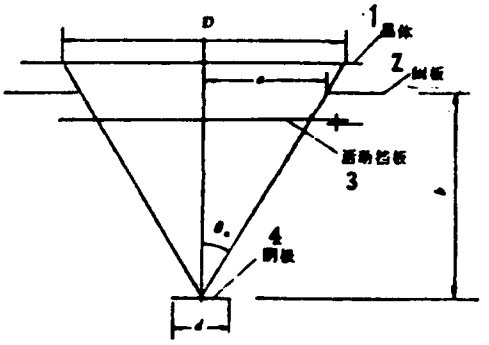

Fig. 2. Schematic diagram of experimental tube for measuring cathode sublimation rate
KEY: 1 - Crystal 2 - Screen plate
3 - Movable back plate
4 - Cathode

The schematic diagram for using the crystal oscillator for measuring the cathode sublimation rate is Fig. 2. In the figure, the purpose of the screen plate is to control the area in which the sublimate is received by the crystal. The purpose of the movable back plate is to open or cut off the passage of the

sublimate at any time in order to precisely control the sublimation time. D is the diameter of the area receiving sublimate by the crystal, d is the cathode diameter, a is the radius of the screen plate hole, and b is the distance between the screen plate and the cathode.

By utilizing the triangular relationship shown in Fig. 2, the molecule number N_0 is replaced by the corresponding mass M_0, the sublimation rate is expressed in v, and only ds at the center of the cathode surface is considered, then from Eq. (2) we obtain:

$$v = \frac{M_0(a^2 + b^2)}{a^2 ds} \tag{3}$$

Considering that the actual sublimation time of measurement is t, the cathode is a cylindrical terminal surface emission cathode with area s (diameter d), then the average sublimation rate of the time period t is as in the following approximate relation:

$$v_t = \frac{M_0^*(a^2 + b^2)}{a^2 st}, \tag{4}$$

In the equation, M_0^* is the mass of the sublimate received by the crystal from actual sublimation of a limited-size cathode.

After the crystal received the sublimate, the variation (decreasing value) df of the oscillation frequency can be expressed as [6,7]:

$$df = -c_f \frac{dm}{F}, \tag{5}$$

In the equation, F is the area in which sublimate was received by the crystal; dm is the mass of the sublimate received by the crystal; c_f is the quality determination sensitivity, which can be experimentally determined according to the actual application situation. Under the above-mentioned conditions of measuring the cathode sublimation rate, this relation is derived. Since the adherent thickness of the sublimate is within the range of hundreds of angstroms or even ten thousand angstroms, df and dm

have a better linear relationship [1,7,8]. Therefore, from Eq. (4) and utilizing Eq. (5), we obtain:

$$v_t = \frac{(a^2 + b^2)}{a^2 c_f}\left(\frac{D}{d}\right)^2 \frac{\Delta f}{t}, \quad (6)$$

in the equation, Δf indicates the decreasing value of oscillation frequency after the crystal received the sublimate.

IV. Inaccuracies of v_t

After the actual cathode is used to replace a flat element, the accuracy thus created can be estimated as follows.

As shown in Fig. 3, if the area in which the sublimate is received is s_0 (2a is the diameter), the cathode area is s (d is the diameter), and two circles are perpendicular to the connecting line oo′ of the circles′ center. Passing through the cathode edge at a point o_1, $o_1o_1' \perp oo_1$, by making $go_1' = o_1'h$, then the area element of the point o_1 is for the sublimation quantity $N_1 = \frac{n\bar{v}}{4}\sin^2\theta_1 ds$ (refer to Eq. (2)) of area s_1 (the diameter is 2(a - r), where r is the cathode radius), for the area element of the point o_1. Actually, the sublimation quantity $N_0' > N_1$ of an area element of the point o_1 that s_0 receives, because other than s_1, the area $(s_0 - s_1)$ already receives the sublimate of the area element of point o_1.

Assume that the sublimation quantity $N_o\left(N_o = \frac{n\bar{v}}{4}\sin^2\theta_o ds\right.$, refer to Eq. (2)), for s_0 by area element at point o, then $N_0 > N_o'$. The following can be proved: refer to Fig. 3, make $fo_1' = o_1'i = co'$, $s_2 = s_0$. Obviously, the sublimation quantity $N_2 = N_0$ to s_2 by area element at point o_1. Since whether s_0 or s_2, for the sublimation of area element at point o_1, the sublimation quantities are the same for the area (the portion marked by hatching in the figure) between fh, however, the included angle ($\theta > \theta_2$) between the partial cone and o_1o_1' corresponding to area I between ef; the included angle $\theta < \theta_2$ between the partial cone and o_1o_1' corresponding to area II between hi. In addition, since the

three-dimensional angle of the area I to o_1 is smaller than the three-dimensional angle of II to o_1, then according to Eq. (1) $N_0'<N_0$ can be obtained. Therefore, $N_1<N_0'<N_0$.

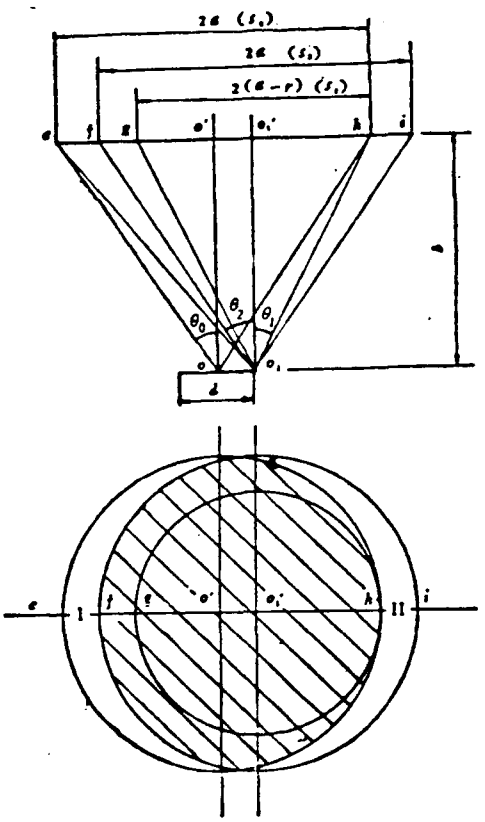

Fig. 3

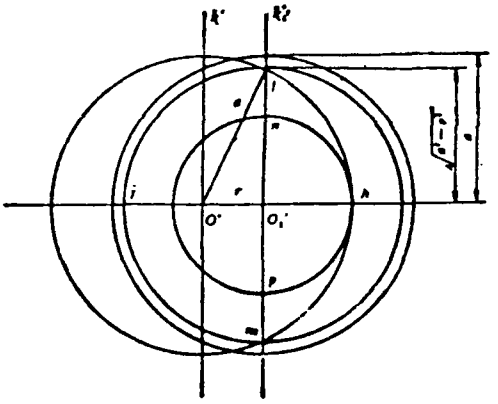

Fig. 4

It can be proved, with the same line of reasoning as above, that to any point o_n between oo_1, there obtains the relation $N_n<N_0^n<N_0$ (N_0^n is the sublimation quantity of area s_0 for the area element of point o_n; N_n is the sublimation quantity of area s_n of area element at point o_n. The diameter of s_n is $2(a - r_n)$,

r_n=oo$_n$). Since $N_1<N_n$, the boundary of inaccuracy is $(N_0-N_1)/N_0$ for N_0 of the sublimation of area elements of various points to s_0.

If we apply Eq. (4) or Eq. (6) to calculate the sublimation rate, actually some number greater than N_1 but smaller than N_0 is considered as N_0, therefore the boundary of inaccuracy of v_t can be expressed as:

$$\frac{M_0 - M_1}{M_0} = \frac{N_0 - N_1}{N_0} = 1 - \frac{1}{\frac{a^2}{a^2 + b^2} + \frac{a^2 b^2}{(a^2 + b^2)(a - r)^2}} \qquad (7)$$

In the following, consideration can be applied to shrink the boundary of inaccuracy. Refer to Fig. 4, by using point o_1' as the center of the circle, and $o_1'l$ as the radius (l is the intersection between $o_1'k_1'$ and the circle by using o' as the center of the circle and a as the radius, among $o_1'k_1'//o'k'$). Then based on Eq. (2) or Eq. (3), the sublimation quantity of area jlno$_1'$pmj for area element of point o_1 (refer to Fig. 3) should be

$\frac{1}{2}\frac{n\bar{v}}{4}\left[\frac{a^2 - r^2}{(a^2 - r^2) + b^2}\right]ds$; similarly, the sublimation quantity of area o_1'nhpo$_1'$ for the area element of point o_1 is $\frac{1}{2}\frac{n\bar{v}}{4}\left[\frac{(a - r)^2}{(a - r)^2 + b^2}\right]ds$.

Thus, it is found that the sublimation quantity N_1' of area jlnhpmj for the area element of point o_1 is

$\frac{1}{2}\frac{n\bar{v}}{4}\left[\frac{a^2 - r^2}{(a^2 - r^2) + b^2} + \frac{(a - r)^2}{(a - r)^2 + b^2}\right]ds$. Therefore, with the above-mentioned analysis, it is found that the boundary of inaccuracy δv_t is further shrunk:

$$\delta v_t = \frac{N_0 - N_1'}{N_0} = 1 - \frac{(a^2 + b^2)}{2a^2}\left[\frac{1}{1 + \frac{b^2}{a^2 - r^2}} + \frac{1}{1 + \frac{b^2}{(a - r)^2}}\right] \qquad (8)$$

Consideration is given to the fact that, during the actual measurement, 0<r<a, therefore, a and b are determined, δv_t increases simply-harmonically with an increase in r. As an

example, if we take a=0.40cm and b=0.50cm, when r is, respectively, 0.15cm, 0.10cm, and 0.05cm, according to Eq. (8), δv_t is, respectively, 29%, 18%, and 8%; if we take a=b=0.50cm, and r is still, respectively, 0.15cm, 0.10cm, and 0.05cm, then according to Eq. (8) δv_t is obtained, respectively, equal to 19%, 12%, and 5%. (The boundary of the actual sublimation rate greater than v_t does not exceed δv_t.)

V. Applications

The above analysis adaptable to the situation in which the crystal is at a relatively close distance from the cathode; at the same time, the effect on crystal temperature of thermal radiation from the cathode is also intensified. The rise in crystal temperature will cause drifting of the oscillation frequency: this is a very key problem in measuring the effect of trace amounts of sublimate. According to reports in reference [2], within the range of -40 to +90^{o}C by cutting crystals for AT with higher thermal stability, the frequency drift is approximately plus or minus 40Hz. When the temperature exceeds 100°C, the frequency drift rises rapidly with increase in temperature. According to the ordinary sublimation rate of the cathode and the geometry of the assumed experimental tube, an estimate is made that the sublimation time required for the frequency to be lowered to 40Hz is approximately several hours. In other words, if we desire that the sublimation time not to be in excess of several hours, the frequency change smaller than 40Hz should be detected. To avoid the frequency drift affecting measurements due to temperature change, in some cases the crystal is to be placed further from the cathode (approximately 6 to 14inches as the distance) and apply cooling to the crystal (maintain the crystal temperature lower than 100^{o}C) [2]. In some cases, two identical crystals are used to operate in identical environments. In one crystal, the sublimate is received; in another crystal, by using a screen as a barrier to most heat

radiation past through, not to receive the cathode sublimate. By measuring the frequency difference of the oscillation frequencies of these two crystals, this is the frequency decrease value [1] caused by the sublimate due to a frequency difference of these two crystals. In the article, frequency measurements are conducted before and after the sublimation to keep the isothermal crystal at the same temperature (at a certain temperature nearing the point of the zero frequency temperature coefficient), the frequency difference is taken as the value of frequency reduction caused by the sublimate thus received. Fig. 5 shows the frequency temperature curve actually measured at different temperatures for an isothermal crystal. The experiment was conducted on a chip of crystal sealed in an experimental tube from which air had been expelled. The marks in the figure show full data measured on the same day (restart of the crystal oscillator). From Fig. 5, if the isothermal temperature during measurement is within the range (approximately between 64.5 and 68.5^{o}C in the figure) of plus or minus 2^{o}C for the measured isothermal temperature at the zero frequency temperature coefficient point, the fluctuation of the crystal isothermal temperature is controlled not to exceed plus or minus 0.5^{o}C, then the range of frequency variation can be smaller than 0.5Hz caused by the fluctuation of isothermal temperature and frequency reappearance (such as temperature increase and decrease for the crystal and restarting of the crystal oscillator). Consideration is given to the results of actual measurement on frequency stability of the crystal oscillator when the oscillation frequency is 5MHz; the range of frequency variation can be smaller than 0.2Hz/24h (not considering the stability of the frequency measurement instruments), after another consideration is given that the crystal received the cathode sublimate on the frequency-time relationship at a certain temperature near the zero frequency temperature coefficient point, the experiment measured that the frequency approximately was reduced by 0.5Hz after 24h; however, when several hours were actually required in

measuring the sublimation rate at a certain temperature. Therefore, the detection change of frequency precise to 1Hz (when the oscillation frequency is 5Mhz) is possible. Now, we consider c_f [6,7], in Eq. (5) we know that the variation of frequency for 1Hz corresponds to barium with a single atomic layer.

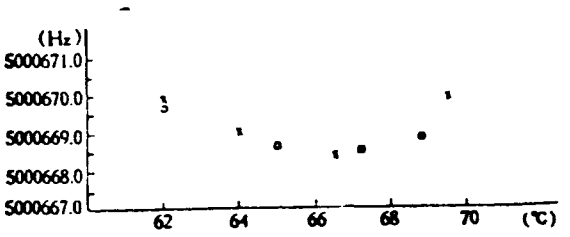

Fig. 5. Crystal oscillator oscillation frequency versus crystal temperature

VI. Conclusions

1. In this method, the sublimate is received within a relatively short distance for a relatively large area, therefore the efficiency of receiving the sublimate is increased, capable of effectively measuring relatively low sublimation rates. By utilizing Eqs. (6) or (4), the sublimation rate can be conveniently calculated for a limited cathode area. The boundary of imprecision for the sublimation rate calculation formula is determined by Eq. (8) (in the derivation, it was assumed that the sublimation rate is the same at various portions of the cathode surface). During applications, requirements can be based to select the values a, b, and r.

2. In this article, measures are taken to overcome the crystal oscillation drift caused by thermal radiation of the cathode, thus basically eliminating the effect on measurements due to frequency drift caused by the crystal temperature rise or the crystal temperature change, thus enhancing the precision of frequency change due to measurement of sublimate. As indicated

by the experiments, when the oscillation frequency is 5MHz, the measured frequency change can be precise to 1Hz.

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REFERENCES

1. "Standard Recommended Practice for Measurement of Sublimation from Thermionic Emitters," ASTM Standards, F53-68 (Reapproved 1978), 274-281.
2. Boyd, W. T., "Measurement of Cathode Sublimation with Commercial Quartz Crystal Oscillators," IEEE Conference Record of 1968 9th Conference on Tube Techniques (1968), 41.
3. Rutledge, W. C. Milch, A. Milch and E. S. Rittner, J. Appl. Phys. 29, 834 (1958).
4. Wang Zhuxi, Tongji Wulixue Daolun [Introduction to Statistical Physics], People's Education Publishing House, 1961, p. 189.
5. Kovalenko, V. F., translated by Information Office of the Xuguang Electron Tube Plant, Rewuli Guocheng He Dianzhenkong Qijian [Thermophysics Processes and Electrovacuum Devices], Defense Industry Publishing House, 1981, p. 100.
6. Compiled by L. Holland and translated by the translation section of the Thin-Film Microelectronics, Bomo Weidianzixue [Thin-Film Microelectronics], Defense Industry Publishing House, 1971, pp. 194-223.
7. Behrndt, K. H. and R. W. Love, Vacuum 12, 1 (1962).
8. Sauerbrey, G., Z. Physik 155, 206 (1959).