DEPOSITION ON A FLAT SUBSTRATE OF GAS MOLECULES EMERGING FROM A CYLINDRICAL CHANNEL WITH AN EVAPORATING INNER SURFACE

V. V. Levdanskii, V. G. Leitsyna, and Khoang Wan Viet

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The article investigates the possibility of controlling the rate of application of matter on a substrate at the expense of varying the temperature along a cylindrical channel, on the surface of which phase transitions take place.

The problem of applying thin layers of matter on various surfaces by the method of gas deposition is very topical [1, 2]. In particular, it is related to producing thin layers having a specified distribution across the thickness or a certain composition required in many technological processes (manufacturing of microelectronic systems, application of optical coatings, protective layers of various kinds, etc.).

By now the external problems of vacuum deposition of matter from a gas phase are fairly completely investigated when the process of deposition is controlled either by varying the position of the particle source with respect to the substrate (for example, by their mutual movement along certain trajectories) or at the expense of geometric parameters of the source proper (the use of extended sources or a system of point sources, arranged in a certain way). To control the process of deposition, use is also made of various masks located between the source and the substrate and changing the spatial distribution of the particle flux incident on the substrate.

Of interest is the investigation of possible methods for controlling the molecular flux entering the substrate which require no change of the source shape or the geometric parameters of the system. One similar method of controlling the deposition process in vacuum application of thin films consists in creating a certain temperature profile at the phase transition surface [3]. In some cases this method of control turns out to be most suitable in producing films of a specified profile, for example, in deposition of matter on the inner surface of a cylindrical channel [4].

The works [3, 5] discussed the possibility of controlling the directivity diagram of particles emerging from a cylindrical channel by varying the temperature distribution along its inner surface, on which evaporation (sublimation)-condensation occurs. The present work deals with the problems of removal of matter from such a channel and its deposition on a substrate located perpendicularly to the channel axis at an arbitrary distance from its outlet.

We will investigate the outflow of gas into a vacuum from a cylindrical channel with length L and radius R, at the distance H from the outlet of which there is a flat substrate (Fig. 1). We assume the flow of the gas in the channel and exterior to it to be free molecular, and the scattering of molecules by the surface to be diffuse. For simplicity we disregard reevaporation of molecules from the substrate (this is valid at a fairly low temperature). We also do not take into account the possibility of the molecules entering the channel from the substrate on their elastic reflection from it. We deal with a step of the process in which the variation in the geometry of the channel due to phase transitions on its inner surface can be disregarded as well as with a simple one-step model of deposition of matter, taking no account of the process of two-dimensional condensation, the effect of the influence of the substrate, etc.

We assume that the temperature varies in a linear manner along the lateral surface of the channel: $T(X) = T_0(1 + AX/L)$, and its relative drop is small, i.e., $|A| << 1$. Such a temperature distribution can be attained, for example, with sufficiently thin channel walls, which are the source of a film-forming evaporating material which is usually characterized by a relatively high thermal conductivity, at the expense of a combination of end and ring heaters in contact with the channel [6].

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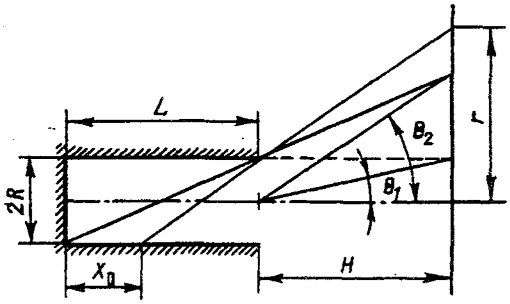

Fig. 1. Geometric diagram of the problem.

The density of the flux of particles evaporating from the surface can be written in the following way [1]:

$$j = \alpha \frac{p_e(T)}{(2\pi mkT)^{1/2}}, \tag{1}$$

where α is the coefficient of evaporation, which is further assumed equal to the condensing (adhesion) coefficient; p_e is the saturated vapor pressure. When the temperature drop is relatively small, p_e can be represented in the form $p_e(T) = p_e(T_0)\exp(sx)$, where $s = (QA)/(kT_0)$. Then in accordance with (1) we have

$$j = \alpha \frac{p_e(T_0)}{(2\pi mkT)^{1/2}} \exp(sx) = j_0 \exp(sx),$$

the value of j_0 being further assumed independent of the coordinate for simplicity.

The thickness of the condensate film δ is determined by the rate of deposition of matter on the substrate v, which is found, taking no account of molecule reevaporation, from the expression

$$n_s v = n_s \frac{d\delta}{dt} = \alpha q,$$

where q is the density of the particle flux entering the substrate; n_s is the density of the condensate material.

Depending on the relation between the channel dimensions and the distance H, two methods for determining q are possible.

If the distance H far exceeds the geometric dimensions of the channel, the particle source formed by it by analogy with [7] can be assumed infinitely small. Then the ratio of the density of the particle flux $q(\theta)$ incident on the substrate at the angle ω to the axis to the value of this quantity for $\theta = 0$, which characterizes the relative film thickness, may be represented in the form [7]

$$f(\theta) = \frac{q(\theta)}{q(0)} = W(\theta)\cos^4\theta, \tag{2}$$

where similarly to [8] the function $W(\theta)$ is expressed as follows:

$$W(\theta) = \begin{cases} \dfrac{4l}{\pi I_0} \displaystyle\int_0^{\frac{\mathrm{tg}\,\theta}{2}} I(x)(1 - l^2\xi^2)^{1/2} d\xi + 1 - \\ - \dfrac{2}{\pi}[\arcsin u + u(1 - u^2)^{1/2}] & \text{при } \theta \leqslant \mathrm{arctg}\,\dfrac{2}{l}, \\ \dfrac{4l}{\pi I_0} \displaystyle\int_0^{1/l} I(x)(1 - l^2\xi^2)^{1/2} d\xi & \text{при } \theta \geqslant \mathrm{arctg}\,\dfrac{2}{l}. \end{cases} \tag{3}$$

Here I(x) and I_0 are the densities of the particle fluxes escaping from the lateral surface of the channel and its bottom; x = X/L is a dimensionless coordinate, $l = L/R$, $\xi = [(1 - x)\tan\theta]/2$, $u = (l\tan\theta/2)$. The terms outside the integral in (3) correspond to straight-transit molecules covering the distance from the bottom to the outlet from the channel without collisions with the lateral surface.

Thus when using relation (2) to find the relative film thickness the channel in fact is replaced by an infinitely small source, which is H distant from the substrate, and the directivity of emerging molecules is calculated from (3), taking into account phase transitions on the lateral surface. From (2) it follows that for finding the absolute value of $q(\alpha)$ it is essential to know the value of q at $\theta = 0$. The latter may be calculated from the density of the particle flux emerging from the channel bottom, situated at the distance H + L from the substrate. In this case a contradiction in determining the distance between the molecule source and the substrate emerges, which is eliminated when H >> L. This is in agreement with the above comment on the applicability of the given approach in cases where H far exceeds the geometric dimensions of the channel.

Turning to the dimensionless particle flux density $\bar{I}(x) = I(x)/j_0$, we obtain for it the integral equation [9]

$$\bar{I}(x) = \exp(sx) + (1-\alpha)\left[\int_0^1 \bar{I}(x_1) K_1(x - x_1)\,dx_1 + \bar{I}_0 K(x)\right], \tag{4}$$

where $\bar{I}_0 = I_0/j_0$ is defined by the relation

$$\bar{I}_0 = 1 + 2l(1-\alpha)\int_0^1 \bar{I}(x) K(x)\,dx. \tag{5}$$

The functions K and K_1 in (4) and (5) characterize the probability of arrival of particles from one surface element at another [10]. Exponential approximation of them [10] makes it possible to obtain an approximate analytical solution of Eq. (4), having for $s \neq \alpha^{1/2}$ the form [9]

$$\bar{I}(x) = a_1\exp(\alpha^{1/2} lx) + a_2\exp(-\alpha^{1/2} lx) + \frac{s^2 - l^2}{s^2 - \alpha l^2}\exp(sx), \tag{6}$$

where the coefficients a_1 and a_2 are found from a system of linear algebraic equations.

From Eq. (4) it is evident that for $\alpha = 1$ there is no necessity to solve this equation, and $\bar{I}(x) = \exp(sx)$; $\bar{I}_0 = 1$ according to (5). In this case from (3) it follows that in the isothermal case (s = 0) the value $\alpha = 1$ corresponds to the equality $W(\theta) = 1$ for all angles θ, i.e., according to (2), we find $f(\theta) = \cos^4\theta$.

When $\alpha \neq 1$ the integration in (3) using (5) and (6) was performed numerically.

We note that the function $f(\theta)$ may be recalculated on f(r), where r is the radial coordinate of a point on the substrate, using the relation $r = H\cdot\tan\theta$.

The approximation of an infinitely small source is inapplicable if the substrate is located fairly close to the outlet from the channel. Here a more general method of calculation should be used when the film thickness distribution is determined not only by the angle at which the particles emerge from the channel but also by the coordinate of the lateral surface element from which they escape. Such an approach was applied in [11-13], where it was assumed that R << H. This assumption enabled the authors to somewhat simplify the calculations but as indicated above in similar situations use could also be made of another method for determining the condensate film thickness, i.e., of the relations (2) and (3). Below in describing the general approach the ratio H/R is assumed arbitrary.

For the dimensionless density of the molecule flux arriving at the substrate from the inner surface of the cylindrical channel by analogy with [11, 13] we have the following expression:

$$\bar{q}_s = \frac{2l^2}{\pi}\int_{x_0}^{1}\int_{\beta}^{\pi}(1 - \rho\cos\varphi)\,\bar{I}(x)\frac{(1 + h - x)\,d\varphi\,dx}{[l^2(1 + h - x)^2 + 1 + \rho^2 - 2\rho\cos\varphi]^2}, \tag{7}$$

where h = H/L, $\rho = r/R$, the values of the lower limits of integration x_0 and β depending on the interval in which the angle θ corresponding to the considered portion of the substrate is located. Introducing the angles θ_1 and θ_2 in the following manner:

$$\mathrm{tg}\,\theta_1 = \frac{R}{H} = \frac{1}{lh}, \quad \mathrm{tg}\,\theta_2 = \frac{(2H + L)R}{LH} = \frac{2h + 1}{lh},$$

we write the relations [11, 13] (see Fig. 1)

$$x_0 = 0,\ \beta = 0 \text{ when } \theta \leqslant \theta_1,$$

$$x_0 = 0,\ \beta = \beta_1 = \arccos\frac{2h-(1-x)(\rho^2-1)}{2\rho h} \text{ when } \theta_1 < \theta < \theta_2,$$

$$x_0 = 1 - \frac{2h}{\rho - 1},\ \beta = \beta_1 \text{ when } \theta \geqslant \theta_2.$$

The expression for the azimuthal angle β_1 in some cross section of the channel $x = \text{const}$ is determined by recognizing that it characterizes the limiting point on the inner surface from which molecules can arrive in the vicinity of the considered point on the substrate (with smaller angles φ, shielding by the channel wall prevents this).

Calculating the integral over φ in (7), we obtain

$$\bar{q}_s = \frac{2l^2}{\pi}\int_{x_0}^{1}(1+h-x)\,\bar{I}(x)\,U(x)\,dx.$$

Here

$$U(x) = \frac{\pi[\Phi(x)-2\rho^2]}{[\Phi^2(x)-4\rho^2]^{3/2}} - \frac{1}{[\Phi^2(x)-4\rho^2]} \times$$

$$\times\left\{\rho[2-\Phi(x)]\frac{\sin\beta}{\Phi(x)-2\rho\cos\beta} + \frac{2[\Phi(x)-2\rho^2]}{[\Phi^2(x)-4\rho^2]^{1/2}} \times \operatorname{arctg}\frac{[\Phi^2(x)-4\rho^2]^{1/2}\operatorname{tg}\frac{\beta}{2}}{\Phi(x)-2\rho}\right\},$$

where

$$\Phi(x) = l^2(1+h-x)^2 + \rho^2 + 1.$$

We pass to evaluating the density of the particle flux entering the substrate from the channel bottom. When $\theta \leq \theta_1$ all molecules escaping from the bottom reach the substrate surface in the region $r \leq R$, and the particle flux density, by recognizing the known analogy between the processes of heat and mass transfer in free molecular flow of gas and those of radiation transfer, will be written in the form [14]

$$\bar{q}_0 = \frac{\bar{I}_0}{2}\left\{1 - \frac{\rho^2 - 1 + l^2(1+h)^2}{([\rho^2+1+l^2(1+h)^2]^2 - 4\rho^2)^{1/2}}\right\}. \tag{8}$$

When $\theta_1 < \theta \leq \theta_2$ partial shielding of the bottom is observed, and the substrate is hit by molecules evaporating from some portion of its surface. In this case the density of such particles is expressed by the relation [11-13]

$$\bar{q}_0 = \frac{2\bar{I}_0}{\pi}\,l^2(1+h)^2\iint_S \frac{\eta\,d\eta\,d\varphi}{[(1+h)^2l^2 + \rho^2 + \eta^2 - 2\rho\eta\cos\varphi]^2}, \tag{9}$$

where S is the (dimensionless) area of the cited portion of the surface. We note that in [11-13] as indicated above it is assumed that $H >> R$ (i.e., $hl >> 1$); therefore in the denominator of the integrand in (9) we can limit ourselves to the first two terms, so the determination of q_0 reduces to calculating the area S. This assumption is lacking in the present work. The limits of integration over η and φ can be found from fairly simple geometric relations, which are not given here. We only point out that these limits are expressed in various ways depending on the difference sign $\theta - \theta_3$, where $\theta_3 = \arctan(1 + 2h)^{1/2}/hl$.

When $\theta > \theta_2$ the molecules escaping from the bottom do not enter the substrate, i.e., for such angles $q_0 = 0$.

The total density of the molecule flux is found from the relation $q = q_s + q_0$, and the relative film thickness is equal to

$$f(\rho) = \frac{\bar{q}(\rho)}{\bar{q}(0)}.$$

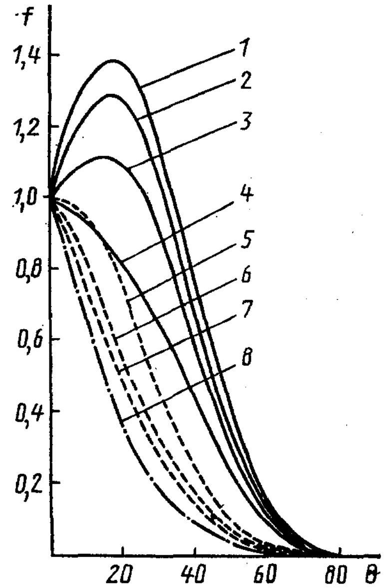

Fig. 2

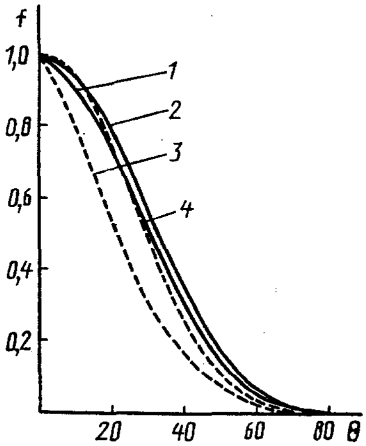

Fig. 3

Fig. 2. Dependence of the relative thickness of the condensate film on the angle for different values of s and h at $a = 1$, $l = 5$. Solid curves correspond to the case $s = 1$, dashed ones to $s = -1$: 1, 7, 8) calculations according to (2) and (3); 2-6) according to (7) and (9); 2) $h = 20$; 3 and 6) 5; 4 and 5) 1. Curve 8 fits the case where a phase transition takes place only at the bottom. θ, deg.

Fig. 3. Dependence of the relative thickness of the condensate film on the angle for different values of s and h for $a = 0.1$, $l = 5$: curves 1 and 3 correspond to calculations according to (2) and (3), curves 2 and 4 according to (7) and (9) at $h = 1$.

Thus, with both methods for describing the process of deposition of molecules their distribution over the substrate depends on the distribution $\bar{I}(x)$ of the density of the particle flux emerging from the lateral channel surface. The function $\bar{I}(x)$ is defined by the dimensionless parameters l, s, and the condensing (evaporation) coefficient α. Therefore these parameters, according to (2) and (3), also affect the dependence of the film thickness on the angle θ when $H \geq R$, and when $H = O(R)$ it depends, in accordance with (7)-(9), on h as well.

When $\alpha = 1$ for an isothermal channel as shown in [9] the flux of emerging particles is equal to the molecule flux evaporating from the bottom. As concerns the distribution of matter over the substrate surface in this case, the numerical calculations, according to (7)-(9), show that it coincides with the distribution of molecule evaporation from a circular disk of radius R located at the distance H from the substrate, i.e., at the location of the channel outlet section.

Numerical calculations are performed for various values of l, s, h, and α by both the methods mentioned above, i.e., according to expressions (2), (3) and by recognizing (7)-(9). They have shown that when $s \leq 0$ the film thickness distribution monotonically decreases with growth of the angle θ, and for $s > 0$, starting from some values of α and h, it is characterized by a maximum (see Figs. 2 and 3). With increasing h the difference between the values of the relative film thickness obtained by different methods decreases, as was to be expected, which is clearly observed in Fig. 2. For a fixed value of h this difference in the case $s < 0$ is smaller than for $s > 0$ (so, when $h = 20$ for $s = -1$ the curves calculated by both methods practically coincide, and for $s = 1$ such a coincidence is observed starting with $h \approx 40$). We note that the distributions f corresponding to positive and negative s (i.e., to the cases where the temperature increases or decreases toward the outlet from the channel) for $\alpha = 1$ differ to a greater extent than for $\alpha = 0.1$. For a fixed α the functions $f(\theta)$ corresponding to drops of the opposite signs differ more distinctly, the larger s is.

Figures 2 and 3 show that for a smaller α the results obtained by different methods become closer with smaller values of h. Figure 2 also gives the function $f(\theta)$ calculated according to (2) and (3) for the case where a phase transition occurs only at the channel bottom. As for the dependence of the character of the distribution $f(\theta)$ on the dimensionless

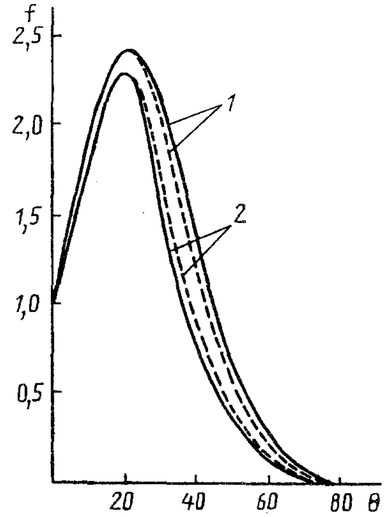

Fig. 4. Dependence of the relative condensate film thickness on the angle in the absence of the bottom for $l = 5$, $h = 5$. Solid curves correspond to $\alpha = 1$, dashed ones to $\alpha = 0.1$: 1) $s = 1$; 2) $s = -1$.

TABLE 1. Values of the Dimensionless Density of the Flux of Molecules Entering the Center of the Substrate for $l = 5$, $h = 5$, and $\alpha = 1$

s	$10^3\bar{q}(0)$	
	channel with bottom	channel without bottom
−1	1.404	0.293
0	1.596	0.480
1	1.981	0.852

channel length $l = L/R$ for fixed H/R, it is insignificant. Calculations have shown that for some values of the parameters (for example, $\alpha = 1$, $l = 5$, $h = 5$, $s = 0.4$) the function f for small θ varies weakly, i.e., the condensate film in the corresponding range of angles is fairly uniform in thickness.

Figure 4 illustrates the distribution of the relative film thickness over the substrate for the channel with no bottom when evaporation is observed only on the lateral channel surface and both ends border the vacuum. In this case in contrast to the one considered earlier the function $f(\theta)$ for the same range of parameters is nonmonotonic for both positive and negative values of s (the maximum being more sharply expressed for $s > 0$), and the dependence on α becomes weaker.

From Table 1, which gives the results of the calculations according to (7)-(9), it is evident that with increasing s the film thickness for $\theta = 0$ increases, i.e., with increasing temperature toward the outlet from the channel q(0) exceeds the corresponding value when the temperature decreases. The same table also gives the data obtained for q(0) in the absence of the bottom. In this case the values of q(0), as was to be expected, decrease in comparison with the corresponding values for the channel with the bottom.

Thus, the work demonstrates that by varying the temperature along a channel with an evaporating inner surface one can affect the distribution of the condensate film thickness over the substrate.

It should be noted that when taking into account the evaporation of molecules from the substrate the rate of deposition of matter is defined by the relation

$$n_s v = \alpha \left[q - \frac{p_e(T)}{(2\pi mkT)^{1/2}} \right]. \tag{10}$$

From (10) it follows that the possibility of controlling the rate of application of matter on the substrate exists both at the expense of varying the temperature along the channel with the evaporating surface (i.e., varying the value of q) and due to formation of the temperature profile T(r) on the deposition surface [4, 15].

In conclusion we point out that the above-considered method of controlling the process of vacuum deposition makes it possible to produce material layers with radial heterogeneity of various kind which can be used, for example, for manufacturing optical filters.

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NOTATION

δ, condensate film thickness; m, mass of molecule; k, Boltzmann constant; Q, heat necessary for evaporation of one particle.

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