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Spatial distribution of gas particles emerging from a nonisothermally heated cylindrical channel with heterogeneous processes on the surface

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Abstract—Spatial distribution is found for a molecular beam emerging from a cylindrical channel into a vacuum with temperature variation along its surface where heterogeneous processes take place. In the case of an evaporating channel surface the behaviour of the functions $f(\theta)$ (relative number of molecules escaping at the angle θ) and $\chi(\theta)$ (relative thickness of condensate film on a substrate) are shown to be determined by the character of temperature variation. Thus, these functions have maxima with temperature increasing towards the exit. The distribution of the concentration of admixture molecules in the film is discussed. The results of calculations for a heterogeneous reaction leading to the disappearance of particles show that, with any character of temperature change along the surface, $f(\theta)$ is a monotonically decreasing function.

INTRODUCTION

The problem of molecular beam control is important for both scientific investigations (for example, when studying the interaction of gas particles with a surface) [1] and different applied areas (the creation of the generators of molecular beams with preset properties, deposition of optical coatings, vacuum deposition, etc.) [2–4].

For the formation of radially symmetric molecular beams, cylindrical channels with impermeable walls are often used where angular distribution of emerging particles is determined only by the ratio of the channel length to its radius [5–10]. In the case of the disappearance of gas particles proper on the channel walls with a heterogeneous chemical reaction the angular distribution depends also on the probability of this process [11]. Here, the control of the spatial distribution of a molecular beam can be effectuated by creating conditions under which the above-mentioned reaction takes place on a part of the lateral channel surface [12]. In sufficiently thin channels the angular distribution of escaping particles experiences the effect of the surface diffusion of molecules adsorbed on the channel walls [13]. In such a case, this distribution can depend on external fields giving rise to the redistribution of molecules adsorbed on the channel surface [14]. In the works mentioned the isothermality of the channel walls was supposed.

One of the ways of controlling the spatial distribution of particles emerging from a channel and, consequently, the process of settling in vacuum depo-

sition of thin films is the use of channels with phase transitions occurring on their internal surface [15, 16].

The present paper is concerned with the analysis of the spatial distribution of a molecular beam outgoing from a channel into vacuum with temperature change along its surface where phase transition or a heterogeneous chemical reaction take place. The regime of flow is assumed to be free molecular.

The deposition of molecules, outgoing from a channel, on a flat substrate is also considered. The influence of the parameters of the problem on the distribution of the thickness of a forming condensate layer is determined. In the case of the presence of admixture molecules in a volume, the behaviour of the concentration of these molecules in a forming layer is analysed.

ANGULAR DISTRIBUTION OF GAS PARTICLES EMERGING FROM A CHANNEL WITH AN EVAPORATING INTERNAL SURFACE

Let us analyse the dependence of the spatial distribution of a molecular (or atomic) beam on the temperature drop along a cylindrical channel (Fig. 1), on the lateral surface and bottom of which evaporation (sublimation)–condensation processes take place, and reveal the role of the parameters that characterize the phase transition.

The evaporation coefficient is considered to be equal to the condensation coefficient and to be independent of temperature. The scattering of particles by the surface is assumed to be diffuse and the temperature to change linearly along the lateral surface

NOMENCLATURE

$f(\theta)$	relative number of particles escaping from a channel at angle θ	r	radial coordinate on substrate
h	dimensionless distance between channel exit section and substrate, H/L	R	channel radius
H	distance between channel exit section and substrate	s	$Q\beta/kT_0$ in Sections 2 and 4, $Q_r\beta/kT_0$ in Section 3
I	density of the flux of particles escaping from lateral surface of a channel	T	temperature
\bar{I}	dimensionless density of the flux of particles escaping from lateral surface of a channel	W	function determined by relation (2)
I_0	density of the flux of particles escaping from bottom	x	dimensionless longitudinal coordinate, X/L
j_0	density of the flux of particles evaporating from surface at $x = 0$	x_0	lower limit of integration in equation (8)
k	Boltzmann constant	X	longitudinal coordinate.
K, K_1	probabilities of arrival of molecules from one surface element to another	Greek symbols	
l	dimensionless channel length, L/R	α	evaporation coefficient
L	channel length	β	coefficient in equation (1)
m	mass of a molecule	γ	probability of gas particle disappearance
n	total molecule density in condensate	θ	angle between direction of particle outflow and channel axis
$N(\theta)$	number of particles escaping from a channel at angle θ	ρ	dimensionless radial coordinate, r/R
p	pressure	ϕ_0	lower limit of integration in equation (8)
q	density of flux of molecules into condensate on substrate	χ	dimensionless thickness of condensate film.
\bar{q}	dimensionless density of the flux of molecules into condensate on substrate	Subscripts	
Q	heat necessary for evaporation of one particle	0	bottom
Q_r	heat of heterogeneous reaction	b	substrate
		e	saturated vapour
		i	isothermal case
		m	maximum number of escaping particles
		s	internal channel surface.

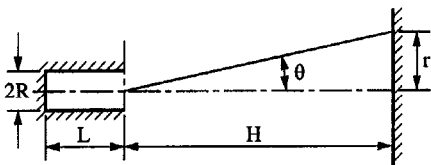


Fig. 1. Geometric diagram of the problem.

$$T(X) = T_0 \left(1 + \beta \frac{X}{L} \right). \quad (1)$$

Such a character of the dependence $T(X)$ can be attained, for example, in the case of sufficiently thin walls of a channel furnishing a source of a film-forming substance (which is usually characterized by a

relatively high thermal conductivity) due to the combination of end and ring heaters being in contact with the channel walls [17]. Moreover, it is assumed that $\beta \ll 1$.

Consider such a stage of the process at which the change in the channel geometry due to phase transitions on its internal surface can be neglected.

The quantity which characterizes the angular distribution of the beam is the ratio of the number of particles, leaving the channel at the angle θ to its axis, to the number of particles, outgoing in the direction of the axis [5]

$$f(\theta) = \frac{N(\theta)}{N(0)} = W(\theta) \cos \theta,$$

where by analogy with ref. [6] the function $W(\theta)$ is expressed as

$$W(\theta) = \frac{4l}{\pi I_0} \int_0^{\tan \theta/2} I(x)(1-l^2 \xi^2)^{1/2} d\xi + 1 - \frac{2}{\pi} \times [\arcsin u + u(1-u^2)^{1/2}] \text{ for } \theta \leq \arctan \frac{2}{l}$$

$$W(\theta) = \frac{4l}{\pi I_0} \int_0^{1/l} I(x)(1-l^2 \xi^2)^{1/2} d\xi \text{ for } \theta > \arctan \frac{2}{l}. \quad (2)$$

Here $x = X/L$ is the dimensionless coordinate reckoned from the bottom assumed to be flat, $l = L/R$, I and I_0 are the densities of the fluxes of particles escaping from the channel lateral surface and bottom, respectively. The quantities ξ and u are defined as $\xi = (1-x) \tan \theta/2$ and $u = l \tan \theta/2$. The terms outside the integral correspond to straight-transit molecules traversing the distance from the bottom to the channel exit without collisions with the walls.

Thus, to determine the angular distribution of molecules emerging from a channel, it is necessary to know the function $I(x)$.

For the dimensionless density of the flux of particles escaping from the lateral surface, when the dimensional density is related to the flux density j_0 of particles evaporating from the surface at $x = 0$, $j_0 = \alpha p_e(T_0)/(2\pi mkT_0)^{1/2}$, the following equation can be written (further the heat of evaporation is assumed to be uniform along the whole channel length) [18]

$$\bar{I}(x) = \frac{p_e(T)}{p_e(T_0)} \left(\frac{T_0}{T} \right)^{1,2} + (1-\alpha) \left[\int_0^1 \bar{I}(x') K_1(|x-x'|) dx' + \bar{I}_0 K(x) \right] \quad (3)$$

where K and K_1 are the functions characterizing the probabilities of the arrival of molecules from one surface element to another, \bar{I}_0 is defined by the relation

$$\bar{I}_0 = 1 + 2l(1-\alpha) \int_0^1 \bar{I}(x) K(x) dx. \quad (4)$$

When $\beta \ll 1$, the quantity $p_e(T)$ can be presented as

$$p_e(T) = p_e(T_0) \exp \{sx\}$$

where $s = Q\beta/kT_0$. Further, for simplicity, in the first right-hand term of equation (3) only the exponential factor is considered to change with temperature, therefore it has the form $\exp \{sx\}$. In such a case, the use of exponential approximation of the functions K , K_1 [5], two-fold differentiation of equation (3) and the combination of the relation obtained with the initial equation yield a differential equation, the solution of which allows one to find an approximate analytical expression for $\bar{I}(x)$ depending on the dimensionless parameters s , l and evaporation (condensation) coefficient α . In accordance with equation (2), the

angular distribution of outgoing particles is determined by the same parameters.

At $\alpha = 1$ there is no need to solve integral equation (3), and $\bar{I}_0 = 1$ according to equation (4). Here, when $|s| \ll 1$, equation (2) gives

$$W(\theta) = \begin{cases} 1 + \frac{2s}{\pi} \left[u(1-u^2)^{1/2} + \arcsin u + \frac{2}{3u} \{ (1-u^2)^{3/2} - 1 \} \right] \\ \text{for } \theta \leq \arctan \frac{2}{l} \\ 1 + s \left(1 - \frac{4}{3\pi u} \right) \text{ for } \theta > \arctan \frac{2}{l}. \end{cases} \quad (5)$$

From equation (5) it follows that in the isothermal case ($s = 0$), when $\alpha = 1$, $W(\theta) = 1$ for all of the angles θ . This corresponds to the satisfaction of the cosine law for the particles escaping from a channel.

For $\alpha \neq 1$ and $s = O(1)$ the angular distribution is found by numerical integration in equation (2) with the use of an approximate analytical solution of equation (3). For the case when evaporation occurs only on the channel bottom, $W(\theta)$ is determined by the relation presented in ref. [5].

The results of calculations for the angular distribution of a molecular beam escaping from a channel with an evaporating surface at $l = 5$, $s = 0$ and 1 are presented in Fig. 2. Curve 2 corresponding to the isothermal case complies with the cosine law. Figure 2 shows that for the fixed values of α and θ the quantity $f(\theta)$ increases with s (for $\alpha = 1$, $|s| \ll 1$ such a charac-

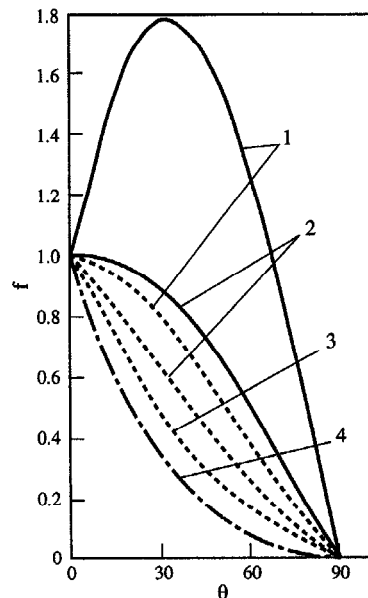


Fig. 2. Spatial distribution of a molecular beam escaping from a channel with evaporation on the surface for $l = 5$. Solid curves correspond to $\alpha = 1$, dashed curves to $\alpha = 0.1$: 1, $s = 1$; 2, $s = 0$; 3, $s = -1$, dashed-dotted curve 4 corresponds to the case of evaporation on the bottom.

ter of the dependence follows from the second relation in set (5), i.e. for any fixed value of α with $s > 0$ (when temperature increases from the channel bottom to the exit) the relative number of particles leaving the channel at a certain angle θ exceeds their number in the isothermal case $f_i(\theta)$, whereas with $s < 0$, $f(\theta) < f_i(\theta)$.

Let us clarify the relationship between $f(\theta)$ and $f_i(\theta)$ for $s < 0$ by taking as an example the simplest case $\alpha = 1$. The farther the part of the surface from the exit section of the channel, the more particles evaporate from it (because of its higher temperature). At the same time, these molecules, escaping at large enough angles, condense on the channel walls, and only the particles emerging at small angles leave the channel. As to the molecules that evaporate from the part of the internal surface of the channel adjacent to its exit section, their quantity here is smaller than in the sections far removed from the exit because of the lower temperature in the exit region. Consequently, for $s < 0$ the ratio of the number of particles escaping at large and small angles, respectively, is smaller than in the case of $s = 0$.

It follows from calculations that for $s \leq 0$ the function $f(\theta)$ is monotonically decreasing, whereas for $s > 0$, starting from a certain value of α , it becomes nonmonotonic. The explanation is that in the latter case the number of molecules leaving the channel at large angles increases (due to both the increase of α and the higher temperature of the surface parts adjacent to the exit). It will be noted that the angle θ_m corresponding to the maximum number of escaping molecules increases with α , as well as with s at a fixed α . Here, when both α and s increase, the value of $f(\theta_m)$ also increases.

Calculations show that as the evaporation coefficient α decreases, the nonisothermality of the surface becomes less noticeable because of the multiple collisions with it of molecules, and the value of $f(\theta)$ approaches that of $f_i(\theta)$ [16].

For comparison, Fig. 2 also contains the function $f_0(\theta)$ describing the distribution of emerging particles in the case when evaporation occurs only on the channel bottom. As seen from the figure, $f_0(\theta) < f_i(\theta)$ for whole range of angles being attributable to the decrease in the evaporation surface area.

As regards the dependence of the spatial distribution on the channel length, then for $\alpha = 1$ and $s = 0$, $f(\theta) = \cos \theta$ in accordance with the foregoing, i.e. such a dependence is absent. When $\alpha \neq 1$, in an isothermal case an increase in the channel length exerts a collimating effect on the flow of emerging molecules resulting in a decrease of $f(\theta)$ [16]. For $s > 0$ with an increasing l the number of molecules, leaving warmer surface parts closer to the exit, increases due to the opposite effect. Thus, for $s < 4$ both the increase in the channel length and the presence of the temperature gradient lead to a decrease of $f(\theta)$, while for $s > 0$ these factors exert the opposite effects, therefore here when l increases the function f can both increase and decrease depending on α and s .

As noted in ref. [19], the character of the angular distribution of molecules emerging from capillaries influences the conditions above a capillary-porous body. Moreover, according to the foregoing, the recession of the bottom of the channel (i.e. increase of l) with the rise in the temperature of the surface receding in the course of evaporation ($s < 0$ and $|s|$ increases) leads to the narrowing of a molecular beam. Note that problems considered can be extended to the calculation of the spatial distribution of radiation emerging from a channel with geometric dimensions substantially exceeding the radiation wavelength [12]. This is possible because of the well-known analogy between a free molecular gas flow and radiation transfer [20].

ANGULAR DISTRIBUTION OF GAS PARTICLES EMERGING FROM A CHANNEL WITH A HETEROGENEOUS REACTION OCCURRING ON ITS INTERNAL SURFACE

Let us find the spatial distribution of particles for a molecular beam passing through a cylindrical channel when a first-order heterogeneous reaction occurs on its internal surface and the surface temperature changes in accordance with equation (1). By analogy with ref. [11], the above-mentioned reaction is understood to be any process with the participation of a wall resulting in a change of the essential properties of the particles with the probability γ independent of the impinging flow (for example, the absorption of particles by the wall, variation in the chemical state of the particles, and so on).

For the first-order reaction the value of γ is proportional to the reaction rate constant [5], therefore the dependence of γ on temperature can be presented as

$$\gamma = \gamma_0 \exp \{-Q_r/kT\}$$

where the pre-exponential factor γ_0 may be considered constant due to a weak dependence on temperature.

The spatial distribution is determined by relation (2) in which the dimensionless density of flux of initial particles leaving the internal surface (the dimensional density is related to the flux density of particles entering the channel) can be found from the following equation

$$\bar{I}(x) = (1 - \gamma_0 \exp \{sx\}) \times \left[\int_0^1 \bar{I}(x') K_1(|x - x'|) dx' + K(x) \right] \quad (6)$$

where $s = Q_r \beta / kT_0$. It should be noted that the equation for the flux density of particles impinging on the unit length of the channel surface in an isothermal case, i.e. for constant γ , was solved in refs. [11] and [21].

In contrast to equation (3), equation (6) includes a multiplier which stands in front of the expression in the square brackets and depends on the coordinate,

while there is a constant multiplier, $(1 - \alpha)$, in equation (3). Therefore, in an isothermal case ($s = 0$) an approximate analytical solution of equation (6) can be obtained as a particular case of the solution of equation (3). However, in the case of a nonisothermal surface of the channel, equation (6) is solved numerically with the use of exact expressions for the functions K and K_1 . For $s = 0$ the results of the solution of this equation obtained by the two above-mentioned methods agree with a good accuracy. The results of the calculations of the spatial distribution for a molecular beam emerging from a channel in the case of a first-order heterogeneous reaction are plotted in Fig. 3. They relate to the case of $l = 10$ and $\gamma_0 = 0.1$, with the values of s being so selected that the value of γ undergoes a 5- and 10-fold increase (decrease) with an increase of x from 0 to 1. The figure also contains a curve which corresponds to the absence of reaction. From Fig. 3 it is seen that, as is to be expected, the presence of reaction narrows the emerging molecular beam, with the collimating effect of the reaction being the greater, the larger the value of s , so that the greatest narrowing of the beam is observed in the case when the temperature, and consequently the probability for a molecule to disappear, increase towards the channel exit. Comparison of Figs. 2 and 3 shows that in the presence of reaction the magnitude of the temperature drop along the channel exerts a much smaller effect on the character of the angular distribution of emerging molecules than in the case of phase transition.

DEPOSITION OF SUBSTANCE FROM A GAS PHASE ON ESCAPE OF MOLECULES FROM A CHANNEL WITH AN EVAPORATING INTERNAL SURFACE

Let us consider the deposition of substance, escaping from a cylindrical channel, on the internal surface and the bottom of which evaporation–condensation processes take place. Suppose that at the distance H from the exit section of the channel there is a flat substrate, which is located perpendicularly to the channel axis and on which condensation of emerging molecules occurs (Fig. 1). For simplicity, the reevaporation of molecules from the substrate is neglected (this is valid at a fairly low surface temperature). The wall temperature is assumed to change linearly in accordance with equation (1).

Replacing, by analogy with ref. [10], the particle source formed by the channel with phase transitions on the internal surface by an infinitely small source with the above-given spatial distribution (2), which is possible when $H \gg R$, it is possible to write the following expression for the relative film thickness (i.e. the thickness of the film related to its value at $\theta = 0$)

$$\chi(\theta) = W(\theta) \cos \theta = f(\theta) \cos^3 \theta. \quad (7)$$

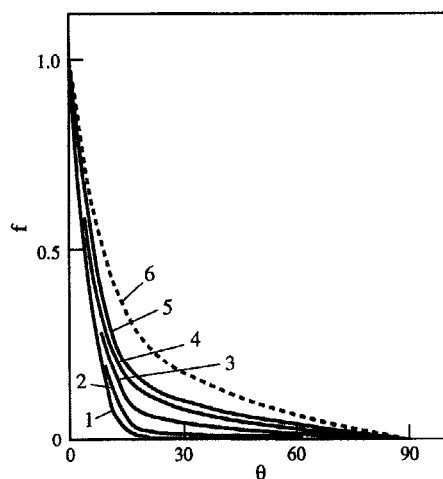


Fig. 3. Spatial distribution of a molecular beam escaping from a channel with heterogeneous reaction on the surface for $l = 10$, $\gamma_0 = 0.1$: 1, $\gamma(1)/\gamma_0 = 10$; 2, $\gamma(1)/\gamma_0 = 5$; 3, $\gamma(1)/\gamma_0 = 1$; 4, $\gamma(1)/\gamma_0 = 0.2$; 5, $\gamma(1)/\gamma_0 = 0.1$, dashed curve 6 corresponds to the absence of reaction.

According to equation (7), the distribution of the film thickness is determined by the behaviour of the function $f(\theta)$. Thus, for $s \leq 0$, when f is a monotonically decreasing function of θ , the quantity $\chi(\theta)$ behaves in the same manner. For $s > 0$, when, beginning from a certain value of α , the function $f(\theta)$ is characterized by a maximum, intervals of relatively uniform distribution of the film thickness $\chi(\theta)$ can be observed. Note that to an infinitely small flat source there corresponds the function $\chi_0(\theta) = \cos^4 \theta$. Thus, for $s > 0$ a nonisothermal channel with phase transitions on its internal surface at certain values of the parameters can be characterized by a more uniform distribution of film thickness than an infinitely small flat source. The approximation of an infinitely small source is inapplicable if the substrate is located fairly close to the channel exit. Here, a more general method of calculation should be used when the film thickness distribution is determined not only by the angle at which 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 refs. [13] and [22] to calculate the spatial distribution of particles emerging from a channel with an impermeable internal surface, where it was assumed that $R \ll H$. This assumption made it possible to somewhat simplify calculations, but, as shown above, a different method of determining the condensate film thickness can be used in such situations, i.e. relation (7). The results of calculations given below are valid for an arbitrary relation between R and H .

For a dimensionless density of the flux of molecules arriving at a substrate from the internal surface of a cylindrical channel the following expression can be written [22]

$$\bar{q}_s = \frac{2l^2}{\pi} \int_0^1 \int_{\phi_0}^{\pi} (1 - \rho \cos \phi) \bar{I}(x) \times \frac{(1+h-x) d\phi dx}{[l^2(1+h-x)^2 + 1 + \rho^2 - 2\rho \cos \phi]^2} \quad (8)$$

where $h = H/L$, $\rho = r/R$. The values of the lower limits of integration in equation (8) depend on the interval in which the angle θ is located which corresponds to the considered portion of the substrate [13, 22, 23].

The total density of the flux of molecules is determined from the relation

$$\bar{q} = \bar{q}_s + \bar{q}_0 \quad (9)$$

where the density of the flux of particles \bar{q}_0 arriving at the substrate from the channel bottom for the arbitrary ratio H/R was calculated in ref. [23]. The relative film thickness is equal to

$$\chi(\rho) = \frac{\bar{q}(\rho)}{\bar{q}(0)}. \quad (10)$$

Thus, with both the methods of describing the process of deposition of molecules, their distribution over the substrate depends on the distribution of the flux density of particles emerging from the lateral channel surface.

As shown in ref. [18], when $\alpha = 1$, the flux of particles emerging in an isothermal channel is equal to the flux of molecules evaporating from the bottom. As concerns the distribution of substance over the substrate surface in this case, calculation according to equation (9) shows that it coincides with the distribution for the evaporation of molecules from a circular disk of radius R located at the place of the channel exit section.

Numerical calculations were performed for various values of l , s , h and α by the above-mentioned methods, i.e. according to expressions (2), (7) and proceeding from equations (9) and (10). They have demonstrated that when $s \leq 0$, the film thickness distribution decreases monotonically with increase of the angle θ , but when $s > 0$, then, beginning from certain values of α and h , it is characterized by a maximum. The results of calculations for $\alpha = 1$ are presented in Fig. 4, where curves 1 and 5 correspond to the approximation of an infinitely small flat source and the remaining curves are found from equations (9) and (10). This figure shows that, as is to be expected, with an increase in h the difference between the values obtained for the relative film thickness by different methods decreases. Here, for a fixed value of h this difference is smaller for $s < 0$ than for $s > 0$ (thus, when $h = 20$ for $s = -1$ the curves calculated by both the methods practically coincide, and for $s = 1$ such a coincidence is observed beginning from $h \approx 40$). Note that where the distributions of χ corresponding to positive and negative values of s (i.e. to the cases when the temperature increases or decreases towards the channel exit) differ the greater, the larger the value of α . For a fixed α the functions $\chi(\theta)$ corresponding to

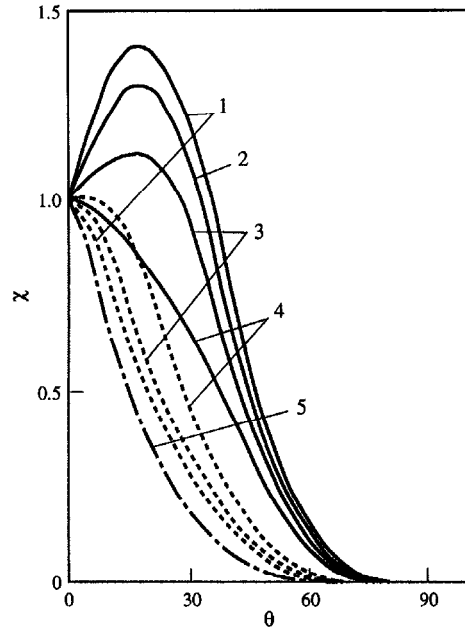


Fig. 4. Dependence of the relative condensate film thickness on the angle with evaporation on the channel surface for $\alpha = 1$, $l = 5$. Solid curves correspond to $s = 1$; dashed curves to $s = -1$; 1, approximation of an infinitely small source; 2, $h = 20$; 3, $h = 5$; 4, $h = 1$, dashed-dotted curve 5 corresponds to evaporation on the bottom.

temperature drops of opposite signs differ more distinctly, the larger $|s|$ value. Calculations show that in the case of a smaller α , the results obtained by different methods get closer for smaller values of h . Figure 4 also contains the function $\chi(\theta)$ calculated according to equations (2) and (7) for the case when phase transition occurs only on the channel bottom. As regards the dependence of the character of the distribution $\chi(\theta)$ on the dimensionless channel length l for a fixed H/R , it is insignificant here.

Calculations according to equations (9) and (10) show that with $\theta = 0$ an increase in s leads to an increase in the film thickness. As is seen from Fig. 1, the function $\chi(\theta)$ can be converted to $\chi(r)$ by means of the relation $r = H \tan \theta$.

Thus, changing the temperature along a channel with an evaporating internal surface, it is possible to influence the character of the distribution of the condensate film thickness over a substrate [16, 24].

Note that the above-considered method of controlling the process of vacuum deposition makes it possible to produce substance layers with various kinds of radial nonuniformity, which can be used, for example, for manufacturing optical filters.

In a number of cases not only the law of substance deposition over the substrate (film thickness distribution) is important, but also the distribution of admixture molecules in the substance layer deposited from a gas phase. It is well-known that even an extremely small concentration of admixture particles

significantly changes the physicochemical properties of the layers obtained (in particular, these problems arise in the production of semiconductor devices). So far as the volume where deposition process occurs practically always contains the molecules of an admixture component and during the condensation of basic substance the trapping of the admixture molecules takes place (including the molecules of the component which is noncondensable at the given values of thermodynamic parameters [25]), it is of interest to investigate the distribution of admixture in the condensate forming over the substrate on the emergence of depositing molecules from a cylindrical channel with evaporating walls. For the concentration of admixture molecules in a solid condensate it is possible to write the following expression [26]

$$c_a = \frac{\alpha_a N_a}{q + J_a} \quad (11)$$

where N_a is density of flux of admixture molecules impinging on the condensate surface (assumed to be independent of r), α_a is sticking coefficient of these molecules, J_a is determined by the relation

$$J_a = n(kT_b/2\pi m_a)^{1/2} \exp\{-Q_a/kT_b\}.$$

In equation (11) the quantity J_a does not depend on the angle θ for constant substrate temperature, and the value of $q(\theta) = \bar{q}(\theta)j_0$ can be evaluated from relation (9). It follows from equation (11) that if $J_a \gg q$ for all the angles θ , the admixture concentration is practically independent of the angle and is equal to $c_a^* = \alpha_a N_a / J_a$. When $q(0) \gg J_a$, then in a certain range of the angles the admixture concentration can be estimated in accordance with equation (11) from the ratio of the particle flux density of an admixture to that of the basic component condensing on a substrate. Here, the dependence of c_a on the angle is determined by the character of the function $q(\theta)$, i.e. for example, in the presence of the maximum for this quantity at the angle θ_m , the admixture concentration has a minimum value for this angle. The value q decreases with increase in θ , and the admixture concentration tends to c_a^* .

CONCLUSIONS

A spatial distribution is found for a molecular beam emerging from a cylindrical channel with temperature change along its surface on which either phase transition or a heterogeneous chemical reaction takes place resulting in the disappearance of particles as such.

The function $f(\theta)$ characterizing the number of molecules escaping from the channel with an evaporating surface at the angle θ is shown to be monotonically decreasing for a constant temperature or a temperature decreasing towards the channel exit. In the case of increasing temperature, the function $f(\theta)$

becomes nonmonotonic and has maximum beginning from a certain value of the evaporation coefficient α .

The presence of a heterogeneous reaction resulting in the disappearance of particles over the channel surface narrows the escaping flux and the largest effect is obtained for the case of temperature increase towards the exit.

The thickness of a condensate film forming on a flat substrate due to the condensation of particles escaping from the channel with an evaporating surface is calculated. It is found that the maximum in the distribution $\chi(\theta)$ with temperature increase towards the exit appears as a consequence of the nonmonotonicity of $f(\theta)$. In the case of the presence of an admixture component in the volume, the character of the concentration distribution of admixture particles in the film is analysed, and it is shown that this also can be a nonmonotonic function of θ .

REFERENCES

1. Yu. N. Lyubotov, Effusion and the processes on the surface, *Usp. Fiz. Nauk* **119**, 641–688 (1976).
2. G. Hass and R. E. Thun, *Physics of Thin Films. Advances in Research and Development*, Vol. 3. Academic Press, New York (1966).
3. L. Holland, *Vacuum Deposition of Thin Films*. Wiley, New York (1956).
4. K. Nanbu and Y. Watanabe, Thickness distribution of films fabricated by the molecular beam epitaxy technique, *Vacuum* **36**, 349–354 (1986).
5. Yu. N. Lyubotov, *Calculation of the Interaction of Molecular Fluxes with Surrounding Vessels*. Izd. Nauka, Moscow (1964).
6. B. B. Dayton, Gas flow patterns at entrance and exit of cylindrical tubes, *Proceedings of the National Symposium on Vacuum Technology*, pp. 5–11. Pergamon Press, London (1957).
7. I. G. Neudachin, B. T. Porodnov and P. E. Suetin, Formation of narrow molecular beams by cylindrical channels, *Zh. Tekh. Fiz.* **42**, 1069–1072 (1972).
8. B. S. Ivanov and V. S. Troitskiy, Concerning the problem of the formation of a directional diagram of molecular beams, *Zh. Tekh. Fiz.* **33**, 494–499 (1963).
9. Yu. N. Grigoriyev, M. Sh. Shavaliyev and V. P. Shapelyev, Mathematical simulation of a tubular source for the molecular-beam epitaxy. In: *Modelling in Mechanics*, Vol. 5, No. 4, pp. 26–37. Izd. SO AN SSSR, Novosibirsk (1991).
10. P. G. Wahlbeck and T. E. Phipps, Effusion—II. Angular number distributions of gaseous cadmium from a right-circular cylindrical orifice into vacuum, *J. Chem. Phys.* **49**, 1603–1608 (1968).
11. A. I. Livshits, I. M. Metter and L. E. Rikenglaz, The angular distribution of a molecular beam at the cylindrical channel exit in the presence of the first-order reaction on the channel walls, *Zh. Tekh. Fiz.* **41**, 368–375 (1971).
12. V. V. Levdansky and Hoang Van Viet, Concerning the control of the angular distribution of a molecular beam escaping from a cylindrical channel. In *Heat and Mass Transfer with Phase and Chemical Transitions*, pp. 143–150. Izd. ITMO AN BSSR, Minsk (1990).
13. W. I. Winterbottom, Vapor–solid interactions and the effusion oven—II. Spatial distribution of an effusion flux, *J. Chem. Phys.* **49**, 105–116 (1968).
14. V. V. Levdansky and Hoang Van Viet, Spatial distribution of energy and mass fluxes escaping from a

- cylindrical channel, Preprint No. 4 of the A. V. Luikov Heat and Mass Transfer Institute, Minsk (1992).
15. V. V. Levdansky, Concerning the control of mass transfer with deposition of thin substance layers by means of precipitation from a gas phase. In *Heat and Mass Transfer in the Technology and Usage of Electronic and Micro-electronic Systems (Proceedings of the International School-Seminar)*, Part 2, pp. 40–47. Izd. ITMO AN BSSR, Minsk (1990).
 16. V. V. Levdansky and V. G. Leitsina, Concerning the influence of nonizothermality of the phase transition surface on the molecular beam formation and film deposition in vacuum, Preprint No. 21 of the A. V. Luikov Heat and Mass Transfer Institute, Minsk (1990).
 17. Yu. Z. Bubnov, M. S. Luriye, F. G. Staros and G. A. Filaretov, *Vacuum Deposition of Films in Quasi-closed Volume*. Izd. Sov. Radio, Moscow (1975).
 18. A. V. Luikov, T. L. Perelman, V. V. Levdansky, V. G. Leitsina and N. V. Pavlyukevich, Theoretical investigation of vapour transfer through a capillary-porous body, *Int. J. Heat Mass Transfer* **17**, 961–970 (1974).
 19. V. V. Levdansky, Phase transitions in a capillary and nonequilibrium heat and mass transfer conditions over a capillary-porous body. In *Heat and Mass Transfer: Physical Foundations and Methods*, pp. 20–21. Izd. ITMO, Minsk (1979).
 20. T. D. Sandry and F. D. Stevenson, Observations on radiation heat transfer and molecular conductance, *J. Chem. Phys.* **55**, 4150–4151 (1971).
 21. F. H. Verhoff and W. Strieder, Numerical studies of Knudsen diffusion and chemical reaction in capillaries of finite length, *Chem. Engng Sci.* **26**, 245–253 (1971).
 22. V. Ruth and J. P. Hirth, The angular distribution of vapor flowing from a Knudsen cell. In *Proceedings of the International Symposium on Condensation and Evaporation of Solids*, pp. 99–145. Gordon and Breach Science Publishers, New York (1962).
 23. V. V. Levdansky, V. G. Leitsina and Hoang Van Viet, Deposition on a flat substrate of gas molecules emerging from a cylindrical channel with an evaporating inner surface, *J. Engng Phys. Thermophys.* **64**, 124–130 (1993).
 24. V. V. Levdansky, V. G. Leitsina and Hoang Van Viet, Vacuum deposition of particles escaping from a cylindrical channel with evaporating walls. In *Kinetic Theory of Transfer Processes with Evaporation and Condensation (Abstracts of Euromech. Colloquium 285)*, pp. 18–21. Minsk (1991).
 25. V. V. Levdansky, Kinetics of the precipitation of a substance from a gas mixture, *J. Engng Phys.* **37**, 1145–1150 (1979).
 26. V. V. Levdansky, Distribution of impurities in layers of substance deposited from the gas phase, *High-purity Substances* **3**, 433–437 (1989).