ABOUT THE INFLUENCE OF PHASE TRANSITIONS ON THE ANGULAR DISTRIBUTION OF PARTICLES EMERGING FROM A CYLINDRICAL CHANNEL

V. V. Levdanskii, N. V. Pavlyukevich, and Khoang Van Viet

UDC 539.198

The influence of evaporation-condensation processes on the walls of a cylindrical channel on the spatial distribution of a molecular beam emerging from it is investigated. The case is considered when a phase transition is realized on a portion of the channel surface.

The problems of formation of molecular (or atomic) beams are topical in many fields of physicochemical investigations [1]. Besides the intensity and composition of a molecular beam it is also important to know its angular (spatial) distribution which, in particular, plays a significant part in obtaining thin layers of matter with a specified distribution in thickness required in many technological processes [2]. As is known [3-6], this distribution depends on many factors, among them the dimensions of a channel, from which the molecular beam emerges as well as heterogeneous processes realized on its walls. In the case when phase transitions take place on the inner surface of the channel, the temperature distribution along the channel proves to be an essential factor affecting the angular distribution of particles, emerging from the channel [7]. It should also be noted that a variation of portions of the channel inner surface differing in the scattering pattern of a molecular beam, for example, there is a heterogeneous process on one of the channel portions, leading to the destruction (or generation) of particles, enables one to also control the angular distribution of the molecular beam going out of the channel [8].

In the proposed work we investigate the angular distribution of particles emerging from a cylindrical channel with a nonisothermal lateral surface, when both the above-mentioned cases are combined. The gas flow regime is assumed free molecular.

We will consider a cylindrical channel with length L and radius R, along the walls of which the temperature drop is prescribed. Let some gas pressure be maintained at one end of the channel, while the condition of vacuum – at the other one. With certain relations of quantities, characterizing the system (the geometric dimensions of the channel, gas pressure, temperature drop as well as physicochemical parameters, describing a phase transition), there may emerge a situation when one portion of the channel surface is impermeable for gas phase particles, whereas on the second portion there is a phase transition. The coordinate of the boundary line between these regions, dependent on the factors mentioned above, is determined in the course of the solution to the problem. The dependence of the angular distribution of particles emerging from the channel on the temperature drop manifests itself through a change in the position of the coordinate of the boundary line between the condensate-covered surface region and the clear one as well as in the intensity of the phase transition along the condensate-covered portion of the surface.

It should be noted that, depending on the direction of a temperature gradient, two cases may take place. If the temperature decreases in the direction of the channel end adjacent to the volume, in which the condition of vacuum is maintained, the condensate-covered portion of the channel may be adjacent to the outlet end of the channel. In this case, for certain values of parameters, there may emerge three zones: a clean surface, a condensate-covered surface, and a clean surface again. With increasing temperature in the direction of the channel end, opening into the vacuum, a condensate-covered surface zone may form at the inlet end of the channel (in this case there exist only two zones). First we will consider the first case.

Academic Scientific Complex A. V. Lykov Institute of Heat and Mass Transfer of the Academy of Sciences of Belarus, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 63, No.3, pp. 326-332, September, 1992. Original article submitted December 18, 1991.

Let the temperature along the channel wall vary by the law

$$T = T_0\left(1 + \frac{T_1 - T_0}{T_0}\frac{X}{L}\right) = T_0(1 + \beta x), \tag{1}$$

where $x = X/L$ is a dimensionless coordinate, directed along the channel axis; T_0 and T_1 ($T_1 < T_0$) are the channel wall temperatures at $x = 0$ and $x = 1$, respectively; further we assume $|\beta| << 1$.

As is well known [9], vapor condensation from a molecular beam of the specified intensity occurs when the substrate temperature is lower than the critical temperature. For simplicity we further assume that the heat of phase transition does not depend on the properties of the substrate and is equal to the evaporation heat of a massive layer of matter; we also do not consider the processes of two-dimensional nucleation preceding condensation. We will find the coordinate of the boundary line x_c between the bare portion of the channel surface and that covered with condensate, supposing that the entire surface from x_c to 1 is covered with condensate (this condition was checked while solving the problem); we assume the layer thickness much smaller than the channel radius. The value of x_c is determined from the equality condition of densities of one-way flows due to the condensation $\alpha_c J^-(x_c)$ and evaporation $j(x_c)$:

$$\alpha_c J^-(x_c) = j(x_c), \tag{2}$$

where $J^-(x_c)$ is the density of the particle flux incident on the channel surface in the vicinity of the point x_c; α_c is the condensing coefficient.

For $J^-(x_c)$ in view of the condensate-covered portion of the surface we have:

$$J^- = N_0 K(x_c) + \int_0^{x_c} I_1(x) K_1(x_c - x)\,dx + \int_{x_c}^{1} I_2(x) K_1(x - x_c)\,dx, \tag{3}$$

where N_0 is the density of the particle flux entering the channel via its end at $x = 0$. The functions $K(x)$ and $K_1(x)$ characterize the probability of a fall of particles from one element of the surface onto the other [1] (we further assume that the particles entering the channel and scattered by the wall are characterized by the Maxwell distribution function). The densities of the particle fluxes $I_1(x)$, $I_2(x)$, escaping from the lateral channel surface in the regions from 0 to x_c and from x_c to 1, respectively, are found from solving the system of equations

$$I_1(x) = N_0 K(x) + \int_0^{x_c} I_1(x') K_1(|x - x'|)\,dx' + \int_{x_c}^{1} I_2(x') K_1(x' - x)\,dx', \tag{4}$$

$$I_2(x) = j(x) + (1 - \alpha_c)\left[N_0 K(x) + \int_0^{x_c} I_1(x') K_1(x - x')\,dx' + \int_{x_c}^{1} I_2(x') K_1(|x - x'|)\,dx'.\right] \tag{5}$$

For $j(x)$ within the framework of a simple one-stage evaporation model the expression follows [10]

$$j(x) = A \exp\left\{-\frac{Q}{kT(x)}\right\}, \tag{6}$$

where Q is the energy of particle evaporation from the condensate surface; T is the condensate surface temperature; k is the Boltzmann constant. The preexponential factor A depends on the specific model employed to describe the kinetics of evaporation [10].

On the other hand, for $j(x)$ one often uses the formula [10]

$$j = \frac{\alpha_e P_e(T)}{(2\pi m k T)^{1/2}}, \tag{7}$$

where $P_e(T)$ is the pressure of saturated vapor of the evaporating matter; α_e is the coefficient of evaporation defined as a ratio of the rate of evaporation into vacuum found experimentally to $P_e(T)/(2\pi mkT)^{1/2}$ [10]. We will note that (7) can also be rewritten in a form analogous to (6); the preexponential factor in this case will include the coefficient of evaporation α_e. Because of a fairly weak dependence on T, for simplicity we will further assume the preexponential factor (both in (6) and (7)) to be independent of temperature.

In approximating K(x) and $K_1(x)$ by the exponential expressions [6], by analogy with [8] one can obtain the analytical solution of the system (4), (5). For the corresponding dimensionless flux densities $\bar{I}_1(x)$ and $\bar{I}_2(x)$, referred to the value of j at temperature T_1, in view of (1), (6) we have

$$\begin{aligned} \bar{I}_1(x) &= a_1 x + a_2, \\ \bar{I}_2(x) &= a_3 \exp\{\sqrt{\alpha_c} l x\} + a_4 \exp\{-\sqrt{\alpha_c} l x\} + \gamma \exp\{sx\}, \end{aligned} \tag{8}$$

where

$$s = \frac{Q(T_1 - T_0)}{kT_1^2}; \quad l = \frac{L}{R}; \quad \gamma = \frac{\exp\{-s\}(s^2 - l^2)}{s^2 - \alpha_c l^2}. \tag{9}$$

The constants a_1, a_2, a_3, and a_4 are found by substituting (8), (9) into the system of equations (4), (5) (nondimensionalized with respect to $j(T_1)$):

$$\begin{gathered} g + \frac{a_1}{l} - a_2 = 0; \\ \left(x_c \exp\{-lx_c\} + \frac{1}{l}\exp\{-lx_c\}\right) a_1 + \exp\{-lx_c\} a_2 - \\ - \frac{\exp\{l(\sqrt{\alpha_c} - 1)\} - \exp\{l(\sqrt{\alpha_c} - 1)x_c\}}{\sqrt{\alpha_c} - 1} a_3 + \\ + \frac{\exp\{-l(\sqrt{\alpha_c} + 1)\} - \exp\{-l(\sqrt{\alpha_c} + 1)x_c\}}{\sqrt{\alpha_c} + 1} a_4 - \\ - \frac{l\gamma}{s - l}[\exp\{s - l\} - \exp\{(s - l)x_c\}] = 0; \\ g + \left(x_c \exp\{lx_c\} - \frac{1}{l}\exp\{lx_c\} + \frac{1}{l}\right) a_1 + (\exp\{lx_c\} - 1) a_2 - \\ - \frac{\exp\{(\sqrt{\alpha_c} + 1) lx_c\}}{\sqrt{\alpha_c} + 1} a_3 + \frac{\exp\{(1 - \sqrt{\alpha_c}) lx_c\}}{\sqrt{\alpha_c} - 1} a_4 - \\ - \frac{l\gamma}{s + l}\exp\{(s + l)x_k\} = 0, \\ \frac{\exp\{\sqrt{\alpha_c} l\}}{\sqrt{\alpha_c} - 1} a_3 - \frac{\exp\{-\sqrt{\alpha_c} l\}}{\sqrt{\alpha_c} + 1} a_4 + \frac{l\gamma}{s - l}\exp\{s\} = 0, \end{gathered} \tag{10}$$

where $g = N_0/j(T_1)$.

Taking the above into account, for the coordinate of the boundary line from (2), (3), and (8)-(10) we obtain the equation

$$\begin{gathered} \frac{s^2 \exp\{(s + \sqrt{\alpha_c} l) x_c - s\}}{s^2 - \alpha_c l^2} + \alpha_c \left[\frac{\exp\{2\sqrt{\alpha_c} l x_c\}}{\alpha_c - 1} + \frac{\exp\{2\sqrt{\alpha_c} l\}}{(\sqrt{\alpha_c} - 1)^2}\right] a_3 + \\ + \frac{\alpha_c \exp\{\sqrt{\alpha_c} l\}}{\sqrt{\alpha_c} - 1} \frac{l(s + l)}{s^2 - \alpha_c l^2} = 0. \end{gathered} \tag{11}$$

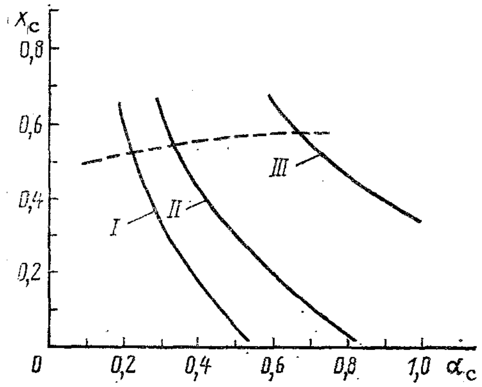

Fig. 1. Coordinate of the boundary line between the condensate-covered and bare zones x_c vs condensing coefficient α_c; $l = 4$, $s = -2.5$: I) $g = 30$; II) 20; III) 10; dashed line refers to the case $\alpha_e = \alpha_c = \alpha$, $\bar{g} = 7$.

Analysis of the numerical solution of Eq. (11) shows that with an increase of the condensing coefficient α_c at fixed values of the other quantities x_c decreases (i.e., the condensate-covered portion of the channel surface increases) (Fig. 1). If the system of equations (4), (5) is referred to $P_e(T_1)\cdot(2\pi mkT_1)^{-1/2}$ rather than to the density of the flux of evaporating molecules at temperature T_1 (as was done above) using expression (7) for j, in (11) there appears one more parameter – the coefficient of evaporation α_e. The dependence of x_c on g in this case can be reduced to the dependence on α_e at a fixed value of $\bar{g} = [N_0/P_e(T_1)](2\pi mkT_1)^{1/2}$. It is interesting to note that if we assume $\alpha_c = \alpha_e = \alpha$ here, then with increasing α the zone covered with condensate decreases (the dashed curve in Fig. 1). With increasing $\alpha = \alpha_e = \alpha_c$ the condensate-covered portion of the surface, on the one hand, evaporates the particles more, but, on the other hand, reflects them less. A decrease in the reflectivity of the condensate in this case leads to a decrease in the particle flux falling onto the surface element in the vicinity of the point x_c and, correspondingly, to a decrease in the condensing particle flux.

We will note that when $\alpha_c = 1$, $I_2 = j$, and the equation for x_c is found directly from (2), (4).

When x_c is found, the angular distribution of the molecular flow $f(\theta)$, characterized by the ratio of the particle flux emerging from the channel at the angle θ to its axis to the particle flux emerging in the direction of the axis, can be written in the form [8]

$$f(\theta) = \left\{1 - \frac{2}{\pi}\left[\arcsin\left(\frac{l\,\mathrm{tg}\,\theta}{2}\right) + \frac{l\,\mathrm{tg}\,\theta}{2}\left(1 - \frac{l^2\,\mathrm{tg}^2\,\theta}{4}\right)^{1/2}\right] + \right.$$
$$+ \int\limits_{\frac{1}{2}\mathrm{tg}\,\theta}^{\frac{1}{2}(1-x_c)\mathrm{tg}\,\theta} \left(-\frac{4l}{\pi}\right)\frac{\bar{I}_1(x)}{g}(1 - l^2t^2)^{1/2}dt +$$
$$\left. + \int\limits_{\frac{1}{2}(1-x_c)\mathrm{tg}\,\theta}^{0} \left(-\frac{4l}{\pi}\right)\frac{\bar{I}_2(x)}{g}(1 - l^2t^2)^{1/2}dt\right\}\cos\theta$$

for $\theta \le \theta_1$, where $\theta_1 = \tan^{-1}(2/l)$ and $t = [(1 - x)\tan\theta]/2$.

For $\theta_1 < \theta \le \theta_2$, where $\theta_2 = \arctan[2/l(1 - x_k)]$ the function $f(\theta)$ takes the form:

$$f(\theta) = \left\{\int\limits_{1/l}^{\frac{1}{2}(1-x_c)\mathrm{tg}\,\theta} \left(-\frac{4l}{\pi}\right)\frac{\bar{I}_1(x)}{g}(1 - l^2t^2)^{1/2}dt + \right.$$
$$\left. + \int\limits_{\frac{1}{2}(1-x_c)\mathrm{tg}\,\theta}^{0} \left(-\frac{4l}{\pi}\right)\frac{\bar{I}_2(x)}{g}(1 - l^2t^2)^{1/2}dt\right\}\cos\theta,$$

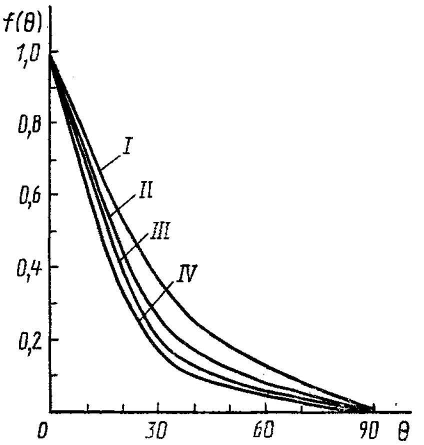

Fig. 2

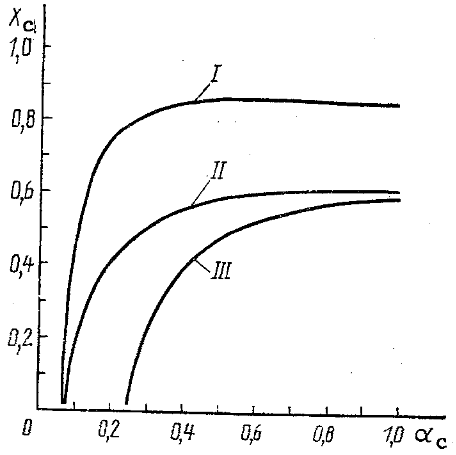

Fig. 3

Fig. 2. Angular distribution of particles emerging from a cylindrical channel vs condensing factor α_c, $l = 4$, $s = -2.5$, $g = 20$: I) impermeable surface, II) $\alpha_c = 0.5$, III) 0.7, IV) 0.8. θ, deg.

Fig. 3. Coordinate of the boundary line between the condensate-covered and bare zones x_c vs condensing coefficient α_c at $l = 4$: I) $s = 0.01$, $g = 20$, II) 1.5 and 20, III) 0.1 and 5.

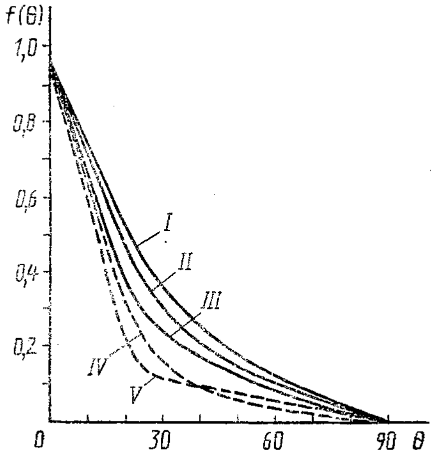

Fig. 4. Angular distribution of particles emerging from a cylindrical channel at the opposite signs of s; $l = 4$: I) impermeable surface, II) $g = 20$, $\alpha_c = 0.4$, $s = -2.5$; III) $g = 20$, $\alpha_c = 0.4$, $s = 2.5$, dashed curves refer to the case of $\alpha_e = \alpha_c = 1$; $\bar{g} = g = 15$; IV) $s = -2.5$; V) $s = 2.5$.

When $\theta > \theta_2$ for $f(\theta)$ we have

$$f(\theta) = \left\{ \int\limits_{1/l}^{0} \left(-\frac{4l}{\pi} \right) \frac{\bar{I}_2(x)}{g} (1 - l^2 t^2)^{1/2} dt \right\} \cos\theta.$$

Figure 2 gives the angular distribution of particles emerging from a cylindrical channel for the case of decreasing temperature towards the outlet end of the channel. For comparison the angular distribution of particles emerging from the channel becomes narrower with an increase of the condensing coefficient α_c. Calculations show that an increase in the temperature drop along the channel leads to a broadening of the beam. The same tendencies also hold for the case of $\alpha_c = \alpha_e = \alpha$ at $\bar{g} = \text{const}$. However, the dependence of f on α in this case is much weaker (the curves are closer to each other).

For the case when the temperature increases towards the outlet end of the channel and the condensate-covered portion of the surface is adjacent to its inlet end (all the quantities being nondimensionalized with respect to $j(T_0)$ and x_c defining the condensate-covered portion of the surface), the value of x_c will increase with increasing α_c and decrease with growing s (Fig. 3). At fairly large values of $g = N_0/j(T_0)$ and small temperature drops the dependence of x_c on α_c can pass through the maximum (curve I in Fig. 3). This is explained by the fact that in the given case there are two competing processes. With growing α_c the probability of condensation of molecules in the vicinity of the point x_c increases, however, the number of molecules entering the channel and reaching the point x_c decreases due to their condensation on the condensate-covered section of the surface.

Figure 4 gives directivity diagrams for the opposite temperature drops at $s = 2.5$ and $s = -2.5$. It is evident from the figure that at the specified parameters the angular distribution proves to be narrower as compared with the angular distribution of particles emerging from the channel with impermeable walls in all cases.

Thus, by varying the temperature along the channel wall (which involves a change in the size of the condensate-covered portion of the lateral surface of the channel) one can affect the directivity diagram of a molecular beam emerging from the channel. This permits, in its turn, the control and distribution of the particle flux, incident onto the substrate at the outlet end of the channel. It should be noted that in the case of a binary mixture of gases, one of the components can condense on the channel walls, while for the other the channel walls (as well as the condensate formed on them) are impermeable, a change in the position of a boundary line between the condensate-covered and clean surfaces involves a change in the relation of component streams emerging from the channel, and in their spatial distribution [11]. Taking into account the known analogy between a free molecular flow of gas and radiation transfer, one can calculate the spatial distribution of the radiation from the channel, which is also connected with the radiation flux distribution across the substrate, located at a distance from the outlet cross section of the channel [12]. This issue can appear to be topical in vacuum application of thin films, since the temperature of the phase transition surface will depend on the energy flux incident onto the given surface.

In conclusion we will make some comments on the methods of refining the solutions obtained by the exponential approximation of the functions $K(x)$ and $K_1(x)$. The given method provides the possibility to obtain approximate solutions of many problems. However, as the ratio of the cylindrical channel length to its radius increases, the exactness of the solution deteriorates. The estimation of the error of the method in question is given in the work [13], in which it is shown that the error in the magnitude of the molecular flow intensity at the end of a long tube does not exceed 12.5%, and by the iteration method, taking the obtained analytical solutions for a zero approximation, one can significantly improve the exactness. The work [14], where the problem of radiation energy transfer within a tube is investigated, presents another method of refinement of the solution, obtained in approximating K and K_1 by approximate expressions, whose employment leads to the results which differ from numerical calculations by no more than 1%. We will also point out that use of several approximating exponents instead of one improves the accuracy of results.

LITERATURE CITED

1. Yu. N. Lyubitov, Usp. Fiz. Nauk, **119**, No. 4, 641-688 (1976).
2. G. Hass and R. E. Tun (eds.), Physics of Thin Films [Russian translation], Vol. 3, Moscow (1968).
3. V. V. Dayton, Proc. Nat. Symp. Tech., Pergamon Press, London–New York–Paris (1957), pp. 5-11.
4. A. I. Livshits, I. M. Metter, and L. E. Rikenglaz, Zh. Tekh. Fiz., **41**, No 2, 368-375 (1971).
5. I. G. Neudachin, B. G. Porodnov, and P. E. Suetin, Zh. Tekh. Fiz., **42**, No 5, 1069-1072 (1972).
6. Yu. N. Lyubitov, Calculation of the Interaction of Molecular Beams with Vessels Fencing Them [in Russian], Moscow (1965), p. 148.
7. V. V. Levdanskii, Heat and Mass Transfer in Technology and Operation of Electron and Microelectron Systems, Proceedings of Int. School-Seminar (Minsk, September 1989), Minsk (1990), Part 2, pp. 142-150.
8. V. V. Levdanskii and Khoang Wan Viet, Heat and Mass Transfer in Phase and Chemical Transitions [in Russian], Minsk (1990), pp. 142-150.
9. Yu. Z. Bubnov, M. S. Lurie, F. G. Staros, and G. A. Filaretov, Vacuum Application of Films in a Quasi-Closed Volume [in Russian], Moscow (1975), p. 160.
10. S. M. Scala and G. L. Vidale, Int. J. Heat Mass Trans., **1**, No. 1, 4-21 (1960).
11. V. V. Levdanskii and Khoang Wan Viet, Kinetic Theory of Transfer Processes in Evaporation and Condensation, Int. School-Seminar (Minsk, September 1989), Minsk (1990), pp. 60-67.
12. V. V. Levdanskii and Khoang Wan Viet, Thermophysics and Hydrogasdynamics-90 [in Russian], Minsk (1990), pp. 6-10.

13. A. I. Ivanovskii, Tr. TsAO, Issue 56, 49-96 (1964).
14. M. Perlmutter and R. Siegel, Heat Transfer, **85**, No. 1, 69-78 (1963).