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CONDENSATION FROM A VAPOR-GAS MIXTURE

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A review of the possible approaches to calculation of vapor condensation from a binary vapor-gas mixture on a surface is presented. Emphasis is paid to justification of the application of molecular-kinetic theory methods for calculation of applied problems. Quantitative estimates for the parameters of the existence in principle of the regimes of one-dimensional stationary condensation are given.

Keywords: *molecular-kinetic theory, condensation, vapor-gas mixture, noncondensable gas, diffusion, Boltzmann equation, condensation choking.*

A. V. Luikov paid great attention to the application of the methods of the molecular-kinetic theory to investigation of the processes of transfer, especially in porous media. In [1], a separate section is devoted to the transfer of vaporous moisture in elementary capillaries. It is noted, in particular, that in macrocapillaries in the presence of a temperature gradient along the length of a capillary there occurs macroscopic motion of the near-wall gas in the direction of the temperature gradient (from the cold end to the hot one), i.e., thermal slipping occurs due to the diluted gas effects. If the ends of the capillary are closed, gas circulation is established in the system. At considerable temperature gradients the thermal slipping of moist air in capillaries is to be taken into account in the total substance transfer. The role of diffusion slipping in the case of a mixture of gases is analogous. In [2], the mechanism of vapor transfer in microcapillaries under nonisothermal conditions in the presence of phase transitions not only on the meniscus of the liquid but also on the surface of the capillary walls is described.

In many heat and mass transfer exchange apparatuses and facilities, transfer processes are realized by liquid evaporation, motion of the vapor formed through the vapor-gas mixture, and condensation of this vapor on the surface whose temperature is maintained lower than the evaporation surface temperature. Phenomena of this kind take place during drying of various bodies, formation of the shielding coatings of the power equipment elements, distillation of substances under lowered pressure (vacuum distillation, chemical deposition of vapor), etc. A noncondensable component present in the investigated region considerably decreases the intensity of condensation [3]. Under certain conditions, even a kind of choking of the interface may occur, i.e., the vapor ceases entirely to condense on it [4–6]. In one of the recent publications devoted to this topic [7] it is noted that investigation of the condensation–evaporation processes in the presence of a noncondensable component is important for development of technologies of storage and transportation of a cryogenic fuel both on the earth and in weightlessness. Consequently, the finding of the conditions for the successful execution of vapor condensation from a vapor-gas mixture is a very urgent problem in the investigation of the corresponding processes of heat and mass transfer and the designing and development of technical facilities of various purpose.

Statement of the Problem. Methods of Solution. Analysis of Results. The methods of the mechanics of a continuous medium developed at the present time adequately describe various flows under condition of a slight departure of the parameters from thermodynamic equilibrium. However, when the processes are in a substantially nonequilibrium state, the problem is to be solved with account for the characteristic molecular-kinetic features of the behavior of a vapor or vapor-gas mixture near the interface.

Works have been published in which vapor condensation from a vapor-gas mixture is considered relative to the study of the processes of heat transfer (see, e.g., [8–12]). It is noted there that the vapor condensation, while exerting an

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appreciable influence on the intensity of heat transfer, represents a complex phenomenon due to the simultaneously occurring heat and mass transfer and the change in the state of aggregation of the substance. Another important factor that influences the heat transfer coefficient is the presence of admixtures of noncondensable gases in the vapor [8, 9, 12–19]. It is noted in these works that in vapor condensation from a vapor-gas mixture the resultant heat transfer coefficient depends on at least two thermal resistances — diffusion resistance and the resistance of the condensate.

In the case of condensation of liquid metal vapors, of great importance is the interphase resistance at the vapor–liquid interface [3]. In [20], an approximate analytical solution of the problem of condensation of pure saturated vapors at low pressures was obtained with the use of the Herz–Knudsen condition at the interface. It is shown that the condensate film thickness in the zero approximation is proportional to $x^{1/3}$.

Film condensation, when a condensed phase is formed on a heat transfer surface, can be observed on various elements of power equaipment.

In addition to this, lately rather wide investigations have been done pertaining to the problem of condensation on solid particles present in a test region or inside a porous body. In [21], on the basis of the model of a "dust gas" the problem of gas flow in a porous body of finite thickness is solved. The integral of collisions in the kinetic equation is replaced by an "external" force resulting from the collective interaction of gas molecules with particles. Simple expressions determining the rate of evaporation from a porous layer, the permeability coefficient, and the density jump at the gas–porous medium interface depending on the Knudsen number Kn ($\mathrm{Kn} = \lambda/L$) are obtained. The expression found for the permeability coefficient allows one to consider the process of gas filtration with adsorbtion in a wide range of Kn values. In [22], on the basis of the method of direct statistical modeling (DSMC) the flow of a two-component gas mixture in a highly porous body was investigated. In calculations the condensation of one component and adsorbtion of the other were taken into account. The distributions of the fluxes of the condensed component and of the adsorbed impurity over the porous layer depth were found, and the efficiencies of the absorption of an impurity in a porous layer and on a plane surface were compared.

A great number of works were devoted to the study of film condensation [9–11, 14, 16]. Usually, in considering a process of such a kind, the conservation equations of a continuous medium written for a vapor-gas medium and a liquid film, as well as the appropriate boundary conditions, are used.

It is traditionally assumed that a vapor is "brought" to the surface by diffusion and that the film surface is impermeable to a gas, i.e., $j_\mathrm{g} = 0$ (the value of the gas mass flux is equal to zero). It should be noted that the diffusion model is strictly admissible for small concentrations of vapor or gas. Using the condition of vanishing of the noncondensable gas mass flux density, i.e., the relation $\rho D \frac{dC_\mathrm{g}}{dx} + \rho C_\mathrm{g} u_x = 0$, we obtain the following expression for the condensed substance mass flux density:

$$j_\mathrm{v} = \frac{\rho D}{L} \ln\left(\frac{1 - C_\mathrm{v}\,(x = L)}{1 - C_\mathrm{v}\,(x = 0)}\right). \tag{1}$$

In experimental work [23] dealing with the drying of medicinal preparations in a drying chamber (with an evaporator and condenser) the validity of Eq. (1) at $\rho_\mathrm{v} \approx \rho_\mathrm{g}$ was confirmed.

It should be emphasized that the use of the purely diffusional approach to condensation or evaporation is not always admissible. It is evident that as a result of vapor evaporation there occurs "displacement" of the noncondensable component (gas) by the vapor moving from the interface. Situations are possible where the gas content in the system is small, and the evaporation intensity suffices for the complete displacement of the gas from the interface. In this case, the relative partial density near the evaporating surface tends to unity, i.e., $C_\mathrm{v}(x = 0) \to 1$. Equation (1) cannot be used in such a situation and, as a consequence, calculations are to be carried out on the basis of other approaches. On the other hand, the other limiting case can also be realized where the pressure of the noncondensable component is much higher than the vapor pressure. In [24], the process of cryogenic pumping of steam through the region filled with gaseous argon was investigated. In the systems considered the argon pressure is in the range $0.5 \cdot 10^{-3}$–$20 \cdot 10^{-3}$ mm Hg, whereas the pressure of steam is several orders of magnitude smaller and amounts to 10^{-8}–10^{-6} mm Hg. The flow of the noncondensable component (argon) in such systems corresponds to a transient or

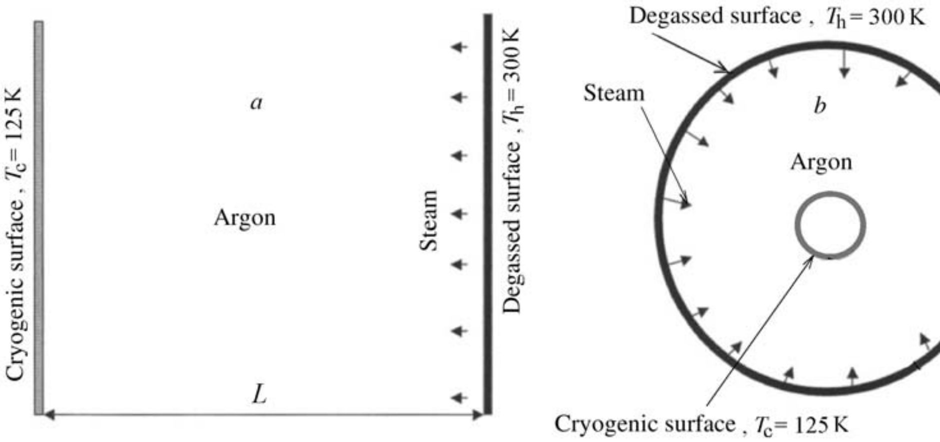

Fig. 1. Cryogenic pumping of steam through the region filled with gaseous argon (the scheme of the problem).

viscous regime. The schematic of the problem is presented in Fig. 1, where a and b represent rectangular and cylindrical geometry, respectively.

From the hot surface (in Fig. 1 it is designated as a degassed one) steam enters the system, with the intensity of the surface "degassing" assumed known and dependent on the material from which the surfaces bounding the test region are made. Usually it is assumed that the steam coming from the surface is brought to the opposite surface (cryogenic surface in Fig. 1) by diffusion and condenses there. However, the use of such a diffusional approach does not allow one to obtain experimentally observed results. For example, if the argon pressure is equal to $P_{\mathrm{Ar}} = 2 \cdot 10^{-3}$ mm Hg, then its density is $\rho_{\mathrm{Ar}} = 4.27 \cdot 10^{-6}$ kg/m^3. As noted above, the steam pressure is several orders of magnitude smaller and amounts to $P_{\mathrm{H_2O}} = 10^{-6}$ mm Hg and, consequently, its density is $\rho_{\mathrm{H_2O}} = 0.96 \cdot 10^{-9}$ kg/m^3. Thus, the density of the mixture is $\rho \approx \rho_{\mathrm{Ar}}$. As a result, $C_{\mathrm{H_2O}} = \rho_{\mathrm{H_2O}}/\rho \approx 0.22 \cdot 10^{-3}$. If $C_{\mathrm{H_2O}} << 1$, Eq. (1) yields

$$j_{\mathrm{v}} = \frac{D}{L} \rho \left(C_{\mathrm{H_2O}}(0) - C_{\mathrm{H_2O}}(L) \right) . \tag{1a}$$

The experimentally observed value of the steam mass flow density is -10^{-8} kg/(m^2·s). From Eq. (1a) one can determine the difference between the relative densities needed for obtaining such a value of the mass flow density:

$$C_{\mathrm{H_2O}}(0) - C_{\mathrm{H_2O}}(L) = \frac{j_{\mathrm{v}} L}{D\rho} . \tag{1b}$$

The test region measures L = 36 in = 0.914 m, and the diffusion coefficient is $D \approx 3 \cdot 10^{-5}$ m^2/s, Further, it follows from Eq. (1b) that $C_{\mathrm{H_2O}}(0) - C_{\mathrm{H_2O}}(L) = -71.35$. Thus, the use of the diffusional approach does not allow one to obtain experimentally observable results.

The calculation of the problem on cryogenic pumping of steam through the argon medium and the derivation of the dependence of pressure on the distance up to the cryogenic panel turned out to be possible only with the use of the molecular-kinetic approach [24].

The basic equation of the molecular-kinetic theory is the Boltzmann equation [25]:

$$\frac{\partial f}{\partial t} + \xi \frac{\partial f}{\partial \mathbf{r}} = J . \tag{2}$$

For the case of a binary gas mixture Eq. (2) is replaced by a system of kinetic equations [25, 18]:

$$\frac{\partial f_a}{\partial t} + \xi \frac{\partial f_a}{\partial \mathbf{r}} = J_{aa} + J_{ab} ,$$

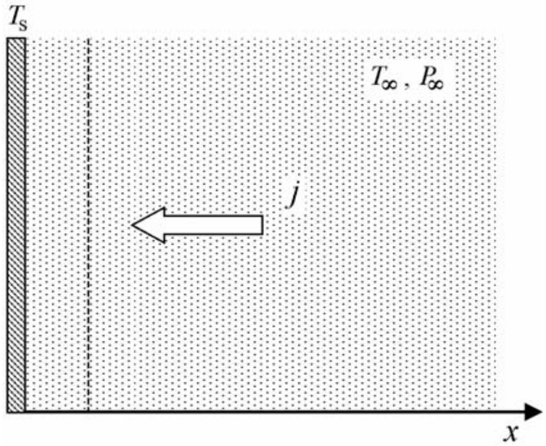

Fig. 2. Schematic of the problem on vapor condensation.

$$\frac{\partial f_b}{\partial t} + \xi \frac{\partial f_b}{\partial \mathbf{r}} = J_{bb} + J_{ba} , \tag{2a}$$

where J_{ij} are the collision integrals that describe the interaction between the molecules of corresponding species (i, j = a, b). From the solution of Eq. (2) or system (2a) one can find the velocity distribution function of vapor (gas) molecules f_i, whose subsequent use makes it possible to find such macroparameters as density, velocity, temperature, pressure, etc. [25].

The Boltzmann equation is a complex integrodifferential equation whose exact analytical solutions can be obtained only for a limited number of real situations. It should be noted that in many cases, in solving problems of hydrodynamics and heat transfer it appears possible to use the equations of a continuous medium, which must be supplemented by appropriate boundary conditions. These conditions should be assigned for all dependent variables, with their form greatly depending on the specific physical problem considered. It is well known [25] that in close vicinity of the interface there exists a thin layer (the so-called Knudsen layer), in which the flow is not described by the Navier–Stokes approximation. To find the solution in the Knudsen layer, where the velocity distribution function of the vapor-gas mixture molecules is not in an equilibrium state, it is necessary to use the methods of the molecular-kinetic theory.

In some works, the process of intense condensation of pure (single component) vapor was investigated in the context of the molecular-kinetic theory [26–30]. Thus, in [27] the condensation of vapor inflowing onto a condensing surface from a semi-infinite space was considered. The problem was investigated in the one-dimensional steady-state statement. The temperature of the interface T_s and the pressure P_s corresponding to this temperature along the saturation line, as well as two quantities out of three [density (or pressure), temperature, vapor flow velocity] far from the condensation surface, were considered known. It was necessary to find the mass flux density and unknown qualities in the gasdynamical flow region, i.e., at a great (as compared to the mean free path of vapor molecules) distance from the inteface. The schematic diagram of the problem is presented in Fig. 2, where the dashed line conventionally designates the boundary of the Knudsen layer located from the interface by several mean free paths. It should be noted that despite the small thickness of this layer, as compared to the macroscopic geometrical dimensions of the problem, for a correct description of vapor behavior beyond the Knudsen layer, one must take into account the specifics of its behavior near the interface.

The results obtained in [27] were approximated by the following relation:

$$j = 1.67 \frac{P_s - P_\infty}{\sqrt{2\pi R T_\infty}} \left\{ 1 + 0.51 \ln \left(\frac{P_\infty}{P_s} \sqrt{\frac{T_s}{T_\infty}} \right) \right\} . \tag{3}$$

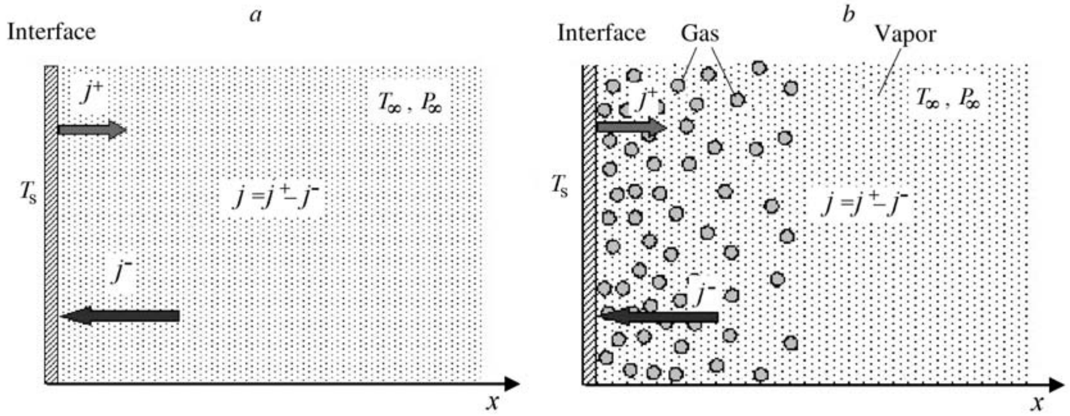

Fig. 3. Influence of the noncondensable component on the process of condensation: a) of a pure vapor; b) from a vapor-gas mixture.

Equation (3) allows one to determine the mass flux density of the condensed substance from the given vapor pressure and temperature. This expression was obtained with regard for the kinetic characteristic features of vapor flow near the interface.

The above situation is complicated considerably when the test region is filled with a mixture of gases. In this case, to determine the macroparameters of both individual components and the mixture as whole it is necessary to solve a system of kinetic equations (2a), each of which includes cross integrals allowing for collisions between components. One of the first works in which a vapor-gas mixture is considered from the standpoint of the molecular kinetic theory seems to be [31]. In [32, 33], on the basis of a model equation for a binary mixture, a problem on evaporation of a substance from one surface and its condensation on another is considered. A small deviation from the state of equilibrium was studied. One of the basic inferences is that the increase in resistance to mass transfer is directly proportional to the amount of the noncondensable substance.

In [34], the authors, while developing the method suggested in [27], considered the problem on vapor condensation in the presence of a noncondensable component for a limited set of mass and concentration ratios of noncondensable and condensable substances.

In [35], condensation of vapor from a volume that contains vapor in a mixture with an inert gas phase is investigated. The case is considered where the condensation rate is much smaller than the thermal velocity of molecules and considerable temperature differences are absent in the system. The estimates presented in that work show that in calculating the process of condensation from a vapor-gas mixture it is permissible to employ quasi-equilibrium boundary conditions if the content of noncondensable impurities in the mixture exceeds a value of the order of the Knudsen number of the system defined as the ratio of the mean free path of molecules λ to the characteristic dimension of the system L. Otherwise, the condensation process is to be considered with regard for the kinetic effects near the phase interface. Such an approach based on correctly posed boundary conditions obtained as a result of the use of the molecular-kinetic theory allows one to avoid the above-noted incorrectness of the limiting transition in Eq. (1) when $C_v(x = 0) \to 1$.

In [7], the problem of recondensation is considered (i.e., where evaporation occurs from one surface and the vapor formed moves through the test section and subsequently condenses on the opposite surface). It is assumed that the vapor formed as a result of evaporation is brought to the condensation surface by diffusion, i.e., the value of the mass flux density of the evaporating (condensing) substance can be found from Eq. (1). Here, to find the mass concentration and temperature of the mixture near the interfaces, kinetic relations obtained as a generalization of results are used [36].

In [4], Baird's Monte Carlo method of direct statistical modeling (DSMC) was adapted to studying the problem of vapor recondensation in the presence of a noncondensable gas. In that work all the molecules were considered mechanically identical, i.e., the ratios of masses and diameters of the molecules of vapor and of a noncondensable substance were equal to unity. According to the results obtained in the work, there exists a situation in which a kind of choking of the evaporation–condensation process occurs because of the presence of a noncondensable component in the region.

From the viewpoint of the molecular-kinetic theory the value of the density of the condensed substance mass flux j is defined as the difference of the densities of two oppositely directed molecular fluxes (see Fig. 3): the flux j^+ of the molecules moving from the interpface (due to evaporation) and the flux j^- of molecules moving to it. With decrease in the temperature of the interface the flux j^+ decreases and, as a consequence, the resultant condensation flux j increases. Thus, an increase in the intensity of the process for a pure substance (Fig. 3a) can be attained by decreasing the temperature of the interface.

On the other hand, if in the region investigated there is a noncondensable gas (Fig. 3b), the main "resistance" to the motion of vapor is due precisely to collisions with the molecules of this gas. In [4], it is noted that "a flux of vapor molecules to the condensation surface is blocked by frequent collisions with noncondensable gas molecules." In this case, even the improvement of conditions for condensation by decreasing the interface temperature does not allow one (in contrast to a pure substance) to attain the required mass flux density of the condensed component [5, 6].

In [6], the approach developed in [27] was extended to condensation in the presence of a noncondensable component by means of a simple addition of the mass conservation equation of the component to a system of conservation equations of mass, momentum, and energy for the whole mixture. Here, as in [27], the boundary conditions were formed proceeding from the nonequilibrium state of vapor near the interface. From the solution of the mass conservation equation of the component, it follows that there exists a limiting value of the gas density near the interface on exceeding which vapor cannot "seep" through this gas. The substitution of the solution of a system of mass, momentum, and energy equations into the expression for such a limiting case, for the first time suggested in [27], allows one to obtain the condition for the existence of one-dimensional steady-state condensation. It is shown in [6] that the ultimately possible regime of condensation is governed by the situation where the relative partial density near the interface tends to zero ($C_{\mathrm{v}}(0) \to 0$). In [5], it is also noted that the vapor density corresponding at saturation to the condensation surface temperature "cannot be negative from the physics of the process." In this case the relative partial density of the gas can be found from the expression

$$C_{\mathrm{g}}^{\lim}(x) = \exp\left[\frac{3j}{4}\int_0^x \frac{dx}{\mu}\right]. \tag{4}$$

From Eq. (4), with regard for [27, 6], the amount of the gas leading to the system choking can be determined from the following expression:

$$\int_{-\infty}^{0} \rho_{\mathrm{g}} d\left(\mathrm{M}_\infty \frac{x}{\lambda_\infty}\right) \le \rho_\infty . \tag{5}$$

It is clear that the approach presented in [6] does not take into account the characteristic features of collisions between the vapor and gas molecules occurring in close vicinity of the interface (in the Knudsen layer). To obtain more accurate estimations of the influence of a noncondensable component on the characteristic features of the condensation process, it is necessary to use a kinetic description. Such a kind of modeling was made in [5]. Just as in [6], it was assumed there that far from the condensation surface only vapor is present. To determine the parameters of a mixture and of each of the components, a model kinetic equation for gas mixtures was used [37]. The interface temperature, the vapor density far from it, and the Mach number of the incoming vapor were considered known. The solution resulted in the distribution of the mixture macroparameters for different initial content of a gas in a system. The authors note that there exist situations where the gas present near the condensation surface impedes the vapor motion, which leads to the cessation of the process of condensation. Such results were obtained for several values of the Mach number. Thus, for example, for $\mathrm{M}_\infty = -0.1$ calculations show that one-dimensional stationary condensation of vapor passing through a vapor-gas mixture becomes impossible if $\int_0^\infty \rho_{\mathrm{g}} d\left(\frac{x}{\lambda_\infty}\right) \ge 9.0\rho_\infty$ even at as low a temperature of the interface as possible. Such a feature is attributable to the blocking influence of the gas near the phase interface, which does not allow vapor to pass in the needed amount to this interface.

Using kinetic calculations [5], one can easily determine the correctness of the estimating procedure in [6]. It follows from (5) that the limiting content of gas is determined by the approximate equality $\int_{-\infty}^{0} \rho_g d\left(M_\infty \frac{x}{\lambda_\infty}\right) \approx \rho_\infty$. For Mach number equal to 0.1 we obtain $\int_{0}^{\infty} \rho_g d\left(\frac{x}{\lambda_\infty}\right) \approx 10.0\rho_\infty$. Thus, an exact kinetic calculation shows that if the initial gas content in the system exceeded $9.0\rho_\infty$, condensation at a rate of $M_\infty = -0.1$ would not occur. On the other hand, the simplified approach points to the fact that condensation would stop if the gas content in the system exceeded $10.0\rho_\infty$. It seems that such an agreement between the results of the precise [5] and approximate [6] approaches is due to the fact that in the latter the boundary conditions for the noncondensable gas are formulated exactly due to the impenetrability of the interface.

Conclusions. An analysis of various approaches to the calculation of condensation from a vapor-gas mixture has shown that in the majority of situations realizable in practice the traditional method based on the diffusion equation and on the condition of the impenetrability of an interface for a gas is valid. However, in the limiting cases of small and high relative partial vapor density near the phase interface the application of the molecular-kinetic theory is required. The authors, with the aid of the approximate method suggested by them, showed that there exist such regions of the values of the gas partial density at which in the one-dimensional steady state complete choking of condensation may occur. These results agree quantitatively with the data of [5] obtained in solving the system of kinetic equations in the entire region considered.

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NOTATION

$C_v = \rho_v/\rho$, $C_g = \rho_g/\rho$, relative partial densities of vapor and gas, respectively; D, coefficient of mutual diffusion, m^2/s; $f_a(\mathbf{r}, t, \xi_a)$, $f_b(\mathbf{r}, t, \xi_b)$, velocity distribution functions of molecules for vapor and gas; j_v, j_g, densities of the mass fluxes of vapor and mass, kg/(m^2·s); j^+, j^-, densities of mass fluxes of molecules moving from the interface and to it, kg/(m^2·s); J, integral of collisions; J_{ij}, collision integrals describing interaction between molecules of corresponding species; Kn, Knudsen number; L, size of the test region, m; M_∞, mach number; P_{H_2O}, P_{Ar}, pressures of steam and argon, Pa; P_s, saturation pressure corresponding to temperature T_s, Pa; P_∞, pressure far from the interface, Pa; $\mathbf{r} = (x, y, z)$, coordinates of molecules in a laboratory coordinate system, m; R, individual gas constant, J/(kg·K); t, time, s; T_s, temperature of the phase interface, K; T_∞, vapor temperature far from the condensation surface, K; u_x, velocity of mixture motion along the x axis, m/s; λ, mean free path of molecules, m; μ, dynamic viscosity of a mixture, kg/(m·s); $\boldsymbol{\xi} = (\xi_x, \xi_y, \xi_z)$, velocity of a molecule in a laboratory coordinate system, m/s; ρ, mixture density, kg/m^3; ρ_v, ρ_g, densities of a vapor and gas, kg/m^3; ρ_{Ar}, ρ_{H_2O}, densities of argon and steam, kg/m^3. Subscripts: c, cool; g, gas; h, hot; lim, limiting; s, at saturation; v, vapor; ∞, far from the interface.

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