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SIMILARITY OF DESUBLIMATION PROCESSES

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Based on a diffusional mathematical model of the processes of desublimation from binary gaseous mixtures, dimensionless similarity numbers are obtained. The effect of these numbers and the parameters contained in them on the thickness and density of a cryogenic precipitate is investigated by numerical methods.

Introduction. The effect exerted by operational parameters of the process of desublimation on the thickness and density of a cryogenic precipitate was investigated experimentally for certain substances, especially for water, rather comprehensively in [1]. However, the formation of the cryogenic precipitate has received little study for the majority of substances that can be isolated from gas mixtures by the method of desublimation.

The main object of the present work was to consider the effect exerted by the substance parameters on the thickness and density of the cryogenic precipitate.

The reason for the formation of a loose layer in the process of desublimation is the same for all of the substances: instability of the plane crystallization front in the presence of a concentration gradient of a desublimating admixture in a boundary layer. This makes it possible to describe the processes of desublimation of various substances by a single mathematical model.

Model. For carrying out the analysis, we can adopt the approach suggested in [2]. With the additional assumption of the linear distribution of temperature over the precipitate layer thickness, the process is described by the system of equations

$$\frac{d\rho_{cr}}{dt} = \frac{MD\left(1 - \dfrac{\rho_{cr}}{\rho_{ice}}\right)}{1.1RT_s h}\left(\frac{d\rho_{sat,ad}}{dT}\right)_s \frac{T_s - T_w}{h}; \tag{1}$$

$$\frac{d}{dt}(\rho_{cr} h) = \frac{\beta M}{RT_f}(p_{ad,f} - p_{ad,sat,s}); \tag{2}$$

$$T_s = \frac{\alpha T_f + \dfrac{\lambda_{cr}}{h} T_w + r\dfrac{d}{dt}(\rho_{cr} h)}{\dfrac{\lambda_{cr}}{h} + \alpha}. \tag{3}$$

To calculate the effective thermal conductivity, we can use averaged relations not associated with specific substances [3, 4]

$$\lambda_{cr} = f(\rho_{cr}, \rho_{ice}, \lambda_{ice}, \lambda_f). \tag{4}$$

The saturated vapor pressure curve can be approximated by the relation

$$p_{sat,ad} = \text{const} \exp\left(-\frac{Mr}{RT}\right). \tag{5}$$

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A comparison made in [4] for the results of a calculation by the model suggested and the model proposed in [2] and experimental data shows that the simplified model is suitable for the analysis of the process.

Separation of Dimensionless Similarity Numbers. Introducing the dimensionless quantities

$$\rho_{cr}^* = \rho_{cr}/\rho_{ice}; \quad h^* = h/h_{max},$$

where

$$h_{max} = \frac{\lambda_{ice}(T_{sat,f} - T_w)}{\alpha(T_f - T_{sat,f})}$$

is the maximum possible thickness of a crystalline layer under the given conditions of heat transfer (in our opinion, this definition of the dimensionless thickness seems to be more convenient for the analysis than $h'^* = h\alpha/\lambda_f$ [5] since in our case the limiting values of the dimensionless density and thickness are normalized to unity);

$$T^* = (T - T_{sat,f})/T_{sat,f}; \quad \lambda_{cr}^* = \lambda_{cr}/\lambda_{ice}; \quad \lambda_f^* = \lambda_f/\lambda_{ice},$$

we transform the system of equations (1)-(5):

$$T_s^* = T_w^* \frac{1 - \frac{\lambda_{cr}^*}{h^*} + K_T\left(1 - \exp\left(K_p \frac{T_s^*}{1 + T_s^*}\right)\right)}{\frac{T_w^*}{T_{cr}^*} - \frac{\lambda_{cr}^*}{h^*}}; \tag{6}$$

$$\frac{d\rho_{cr}^*}{dt^*} = K_p \lambda_f^* \frac{(1 - \rho_{cr}^*)(T_s^* - T_w^*)(T_f^*)^2}{1.1(T_w^*)^2 (h^*)^2 (T_s^* + 1)^3} \exp\left(K_p \frac{T_s^*}{1 + T_s^*}\right); \tag{7}$$

$$\frac{d(\rho_{cr}^* h^*)}{dt^*} = -\frac{T_f^*}{T_w^*(T_f^* + 1)}\left(1 - \exp\left(K_p \frac{T_s^*}{1 + T_s^*}\right)\right); \tag{8}$$

$$\lambda_{cr}^* = f(\rho_{cr}^*, \lambda_f^*). \tag{9}$$

In the resulting system of equations all the characteristics of the substance are concentrated in four dimensionless numbers:

$$t^* = \frac{\alpha^2 DMp_{ad,f} t}{\rho_{ice} RT_{sat} \lambda_f \lambda_{ice}}$$

is the dimensionless time;

$$K_T = \frac{rDp_{ad,f}M}{R\lambda_f T_f (T_f - T_{sat,f})} = \frac{rD\rho_f}{\lambda_f T_f^* T_{sat,f}}$$

characterizes the ratio of heat of the phase transition to the convective component of heat inflow;

$$K_p = \frac{Mr}{RT_{sat,f}}$$

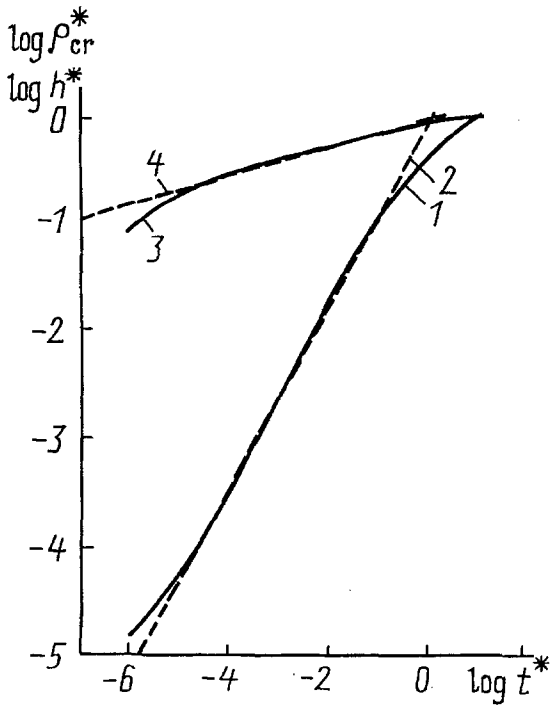

Fig. 1. Dependence of the dimensionless thickness and density of a cryogenic precipitate on the dimensionless time ($T^*_w = -0.2$; $T^*_f = 0.2$; $K_T = 0.5$; $K_p = 15$; $\lambda^*_f = 0.05$); 1, h'^*, calculation by the dimensionless model; 2, $h^* \approx 0.8 (t^*)^{0.86}$; 3, ρ^*_{cr}, calculation by the dimensionless model; 4, $\rho^*_{cr} \approx 0.8(t^*)^{0.13}$.

characterizes the ratio of the energy barrier of the phase transition to the energy of the thermal motion of the molecules;

$$\lambda^*_f = \lambda_f/\lambda_{ice}$$

is the ratio of the thermal conductivity coefficients of the gas phase and the dendritic structure skeleton.

The dimensionless parameters T^*_w and T^*_{cr} are determined exclusively by the conditions of the occurrence of the process.

Analysis of the Dimensionless Numbers. The dependence of h^* and ρ^*_{cr} on t^* is rather well approximated by functions of the form

$$h^* \sim (t^*)^a, \quad \rho^*_{cr} \sim (t^*)^b,$$

where $a < 1$ and $b < 1$ are constants determined by the numbers K_T, K_p, λ^*_f, T^*_w, T^*_f (Fig. 1). At very high values of t^* the thickness and the density approach limiting values, and this dependence is violated.

An increase in the number K_T leads to an increase in the density and a decrease in the relative thickness of the cryogenic precipitate. In many cases that are important in practice, when $\rho_{ad,f}$ is small and the flow temperature greatly exceeds the dew point ($T_f > T_{sat,f}$), the number K_T is small and its effect can be neglected.

An increase in the number λ^*_f leads to an increase in the density (Fig. 2). Consequently, to obtain a denser cryogenic precipitate of a given substance in freezers, a noncondensing gas with a high specific thermal conductivity can be used. When it is necessary to obtain a loose crumbling cryogenic precipitate, it is better to use a noncondensing gas with a low specific thermal conductivity.

In the beginning of the process the dependence of h^* on λ^*_f is a monotonically decreasing one. At a large value of the dimensionless time (of the order of 1), when h^* approaches its limiting value, the dependence of h^* on λ^*_f becomes an increasing one.

TABLE 1. Estimate of the Minimum Value of K_p from the Data of [6]

Condensing substance	Minimal value of K_p	Condensing substance	Minimal value of K_p
H_2	7.6	N_2	10.6
Ne	8.6	O_2	15.1
CH_4	8.9	H_2O	15.9
Ar	9.2	CO_2	19.7
Xe	9.4	C_3H_6	25.4
Kr	9.5	C_3H_6	26.6

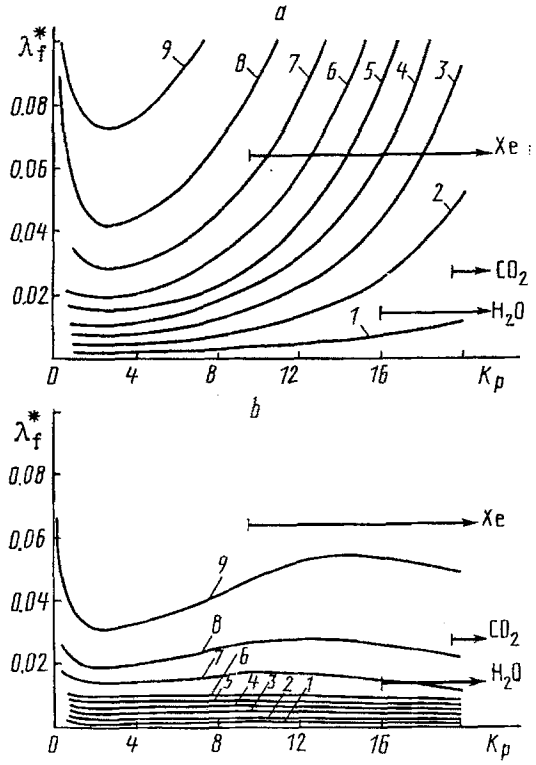

Fig. 2. Levels of the dimensionless density of a cryogenic precipitate as functions of the numbers K_p and λ_f^* for the instants of time $t^* = 10^{-3}$ (a) and $t^* = 1$ (b) ($T_w^* = -0.2$, $T_f^* = 0.2$; $K_T = 0.5$); 1, $\rho_{cr}^* = 0.1$; 2, 0.2; 3, 0.3; 4, 0.4; 5, 0.5; 6, 0.6; 0.7; 8, 0.8; 9, 0.9. Arrows denote admissible regiond of values of K_T and λ_f^* for certain substances.

The dependence of ρ_{cr}^* on the number K_p has two extrema (Fig. 2). For each substance the range of values of K_p is bounded from below (see Table 1) by the melting point. At smaller values of K_p the surface of the cryogenic precipitate can fuse, and the analysis based on the diffusional model is inapplicable.

None of the substances considered falls into the region of the first extremum of the dependence of ρ_{cr}^* on K_p, and therefore for them the increase in K_p at the start of the process leads to a decrease in ρ_{cr}^* (Fig. 2a). At a rather large value of the dimensionless time, the second extremum appears: starting from a certain value, an increase in K_p leads to an insignificant increase in ρ_{cr}^* (Fig. 2b).

The dependence of h_{cr}^* on K_p is a monotonically increasing one. Only at values of t^* of the order of 1 are very insignificant errors noted in the results of the calculations.

Use of the Dimensionless Numbers. The proposed dimensionless numbers permit one to follow the effect of certain thermophysical properties of substances on the process of desublimation. Thus, the quantities α^2 and $1/\rho_{ice}$ enter only into the dimensionless time and therefore exert the same effect on h^* and ρ_{cr}^* as the time:

$$h^* \sim (\alpha)^{2a} \sim (\rho_{ice})^{(-a)}, \quad \rho_{cr}^* \sim (\alpha)^{2b} \sim (\rho_{ice})^{(-b)}.$$

The diffusion coefficient of the admixture enters in the numbers K_T and t^*. In cases where it is possible to neglect the effect of the number K_T ($K_T \approx 0$), we obtain $h^* \sim D^a$, $\rho_{cr}^* \sim D^b$.

Knowing the dynamics of the build-up of the cryogenic precipitate from one experiment or calculation and having determined the coefficients a and b, we may predict the behavior of the process under other conditions of heat transfer or diffusion and for the substances with a different density of the crystal.

The present approach permits one to establish the dependence of h^* and ρ_{cr}^* on the mixture pressure, but the result obtained does not correspond to experimental data due to the incompleteness of the mathematical reference model, which, in particular, does not describe convective transfer in the interior of the cryogenic precipitate and the specific features of the formation of surface layer. Potential possibilities for accounting for the factors indicated are afforded by a nonstationary mathematical model that considers the distribution of the density over the cryogenic precipitate thickness [4].

By all means, the set of dimensionless numbers suggested in the present work and based on a specific mathematical model is not complete and should be extended for a more detailed analysis of the process.

CONCLUSIONS

1. Based on a diffusional mathematical model of the process of desublimation, the parameters determined by the properties of a substance have been grouped into four dimensionless similarity numbers.

2. Using the method of finite differences, the effect of dimensionless numbers on the thickness and density of a cryogenic precipitate has been investigated.

3. Functional dependences of the thickness and density of a cryogenic precipitate on the time, density of crystals, and heat transfer and diffusion coefficients have been obtained.

4. To obtain a denser cryogenic precipitate of a specific substance, we suggested that a noncondensing gas with a higher specific thermal conductivity be used.

NOTATION

D, diffusion coefficient of admixture; h, thickness of cryogenic precipitate; M, mass of admixture mole; p, pressure; R, universal gas constant; t, time; T, temperature; α, heat transfer coefficient; β, mass transfer coefficient; λ, coefficient of specific thermal conductivity; ρ, density; r, heat of phase transition of admixture. Subscripts: cr, cryogenic precipitate; sat, saturation; f, flow; ad, admixture; w, wall; ice, crystals; s, surface of cryogenic precipitate. An asterisk denotes relative (dimensionless) values; a tilde indicates direct proportionality.

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