TRANSIENT HEAT AND MASS TRANSFER IN THE TWO-PHASE SYSTEM: SUBLIMING SOLID-VAPOUR-GAS MIXTURE

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Abstract—Transient heat and mass transfer in the two-phase system consisting of a subliming solid and a vapour-gas mixture is analysed. The aim of the work is to determine the temperature of the sublimation surface of a solid substance, evaporated from a plate into a layer of inert gas for the case when the plate temperature is a function of time. The mathematical description of the system is verified experimentally by determining the temperature profiles in a layer of solid naphthalene in a naphthalene—helium system. The good agreement found between the calculated and measured temperature profiles justifies the approximations used and verifies the results of an earlier analysis of unsteady state heat and mass transfer in the thermal diffusion cloud chamber.

NOMENCLATURE

 A_{vg} , A_{gv} , constant in the Wassiljewa equation;

 C_p , heat capacity at constant pressure;

 C_{ν} , heat capacity at constant volume;

c, mole concentration;

 D_{vg} , coefficient of binary diffusion, $D_{vg}^0 R T^{s+1}/P$;

e, energy flux relative to mass average velocity;

H, enthalpy;

h, height of vapour-gas layer;

J, molar flux relative to mass average velocity;

k, thermal conductivity;

L, chamber height;

N, molar flux relative to stationary coordinates;

n, number of moles;

P, pressure;

q, energy flux relative to stationary coordinates;

R, gas constant;

r, heating rate;

s, constant;

T, temperature;

t, time;

U, internal energy;

v, velocity;

x, mole fraction;

Z, dimensionless length coordinate, z/L;

z, length coordinate.

Greek symbols

 α , thermal diffusion factor;

 ρ , density.

Superscripts

, related to the unit of mass;

–, partial molar quantity.

Subscripts

v, g, quantity in the binary system.

INTRODUCTION

IN MANY technical processes we encounter the problem of transient heat transfer in a composite system. In

many cases, this process is combined with a phase change at the boundary. It is only natural to try to develop a suitable analytical or numerical method for evaluating the temperature distribution and the position of the phase boundary as functions of time [1-8]. In this contribution, we tackle the problem of transient heat and mass transfer in the two-phase system consisting of a subliming substance and a vapour-gas mixture when the temperature of the outside surface of the solid phase changes with time. This particular problem is connected with an experimental study of nucleation in supersaturated vapour of solids in the thermal diffusion cloud chamber and its aim is to determine the temperature of the subliming surface which forms the boundary condition for the evaluation of the supersaturation profile in the vapourgas mixture [9].

SYSTEM DESCRIPTION

The system examined is depicted in Fig. 1. It consists of a layer of a subliming solid substance, placed on the top plate of the chamber, and a layer of a vapour-gas mixture. At the initial stage of measurements, the system is in the steady state: during measurements the temperature of the upper plate provided with the layer of the subliming solid increases with time, while the temperature of the bottom plate is constant. In the system description it is assumed that heat and mass fluxes are unidimensional and that the vapour-gas mixture behaves ideally. As the total time of measurement is generally short, and the mass flux is small, it is assumed further that the position of the boundary is constant during the measurements. For the

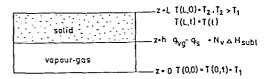


Fig. 1. The coordinate system.

same reason, it is also assumed that the thickness of the layer of the solid substance, forming on the surface of the cooled plate by condensation, is negligible. The temperature of the condensing surface is then equal to the temperature of the plate surface. By v we denote the vapour of the subliming solid and by g the inert gas.

The chamber represents a closed two-component system where the only external force is gravity and the driving forces of heat and mass transfer are the concentration and temperature gradients. For such a case the heat and mass fluxes in the system are described by [10]

$$q_s = -k_s(dT/dz), \quad h < z \le L, \tag{1}$$

$$q_{\mathrm{vg}} = -k_{\mathrm{vg}}(\mathrm{d}T/\mathrm{d}z) + N_{\mathrm{v}}\bar{H}_{\mathrm{v}} + N_{\mathrm{g}}\bar{H}_{\mathrm{g}}$$

$$+\alpha RT(x_gN_v-x_vN_g), \quad 0 \le z < h,$$
 (2)

$$N_{\rm v} = -(c/x_{\rm g})D_{\rm vg}[({\rm d}x_{\rm v}/{\rm d}z) + \alpha x_{\rm v}x_{\rm g}({\rm d}\ln T/{\rm d}z)]$$

$$+x_{\mathbf{v}}N_{\mathbf{g}}/x_{\mathbf{g}}, \quad 0 \leqslant z < h.$$
 (3)

In steady state, when vapour diffuses through the stationary inert gas $g(N_g=0)$, equations (1)–(3) can be adjusted for the calculation of temperature and partial pressure profiles to give

$$dT/dz = -q_s/k_s, \quad h < z \le L, \tag{4}$$

$$dT/dz = k_{vg}^{-1} \{ -q_{vg} + N_v [\bar{H}_v + \alpha RT(P - P_v)/P] \},$$

$$0 \leqslant z < h, \quad (5)$$

$$dP_{\rm v}/dz = \alpha P_{\rm v}(P_{\rm v} - P)/TP(dT/dz)$$

$$+(P_{v}-P)N_{v}/T^{s}D_{vs}^{0}, \quad 0 \le z < h, \quad (6)$$

with the following boundary conditions (cf. Fig. 1):

$$T(0) = T_1, \quad P_{v}(0) = P_{v,eq}(T_1), \quad z = 0,$$

$$q_{vg} - q_s = N_v \Delta H_{subl}, \quad P_{v}(h) = P_{v,eq}[T(h)], \quad z = h,$$

$$T(L) = T_2, \quad T_2 > T_1, \quad z = L. \tag{7}$$

To describe the temperature and partial pressure profiles during heating we may start off by considering equation (3), the equation of continuity of the component v, and the energy equation [11]

$$\rho \hat{C}_{v}[\partial T/\partial t + v(\partial T\partial z)]$$

$$= -\partial c/\partial z - P(\partial v/\partial z) + \bar{U}_{v}(\partial J_{v}/\partial z) + \bar{U}_{g}(\partial J_{g}/\partial Z) \quad (8)$$

where the following relation is valid for the energy fluxes q and e:

$$e = q - v(c_{\mathbf{v}}\bar{H}_{\mathbf{v}} + c_{\mathbf{v}}\bar{H}_{\mathbf{v}}). \tag{9}$$

Assuming ideal behaviour and rearranging the results for the profiles in ref. [9] we have

$$\partial T/\partial t = A(\partial^2 T/\partial z^2) + B(\partial T/\partial z), \quad h < z \le L, \quad (10)$$

$$\partial T/\partial t = C(\partial^2 T/\partial z^2) + D(\partial T/\partial z) + E, \quad 0 \le z < h,$$
(11)

$$\partial P_{\mathbf{v}}/\partial t = F(\partial^2 P_{\mathbf{v}}/\partial z^2) + G(\partial P_{\mathbf{v}}/\partial z) + H, \quad 0 \le z < h$$
(12)

where the coefficients A-H are generally functions of state variables, compositions, and their derivatives (see

Appendix). The initial conditions for equations (10)-(12) are given by the steady state solution. The boundary conditions (cf. Fig. 1) are given by

$$T(0,t) = T_{1}, \quad P_{v}(0,t) = P_{v,eq}(T_{1}), \quad z = 0, \quad t > 0,$$

$$q_{vg}(h,t) - q_{s}(h,t) = N_{v}(h,t)\Delta H_{subl},$$

$$P_{v}(h,t) = P_{v,eq}[T(h,t)], \quad z = h, \quad t > 0,$$

$$T(L,t) = f(t), \quad z = L, \quad t > 0.$$
(13)

Mass fluxes are given by the definition relation (3) and the equation of continuity for the component g,

$$\partial c_{\mathbf{g}}/\partial t = -(\partial N_{\mathbf{g}}/\partial z), \quad 0 \le z < h$$
 (14)

with the boundary condition $N_g(h,t) = N_g(0,t) = 0$. The total pressure of the vapour-gas mixture follows from the balance of the total number of moles,

$$n(t) = n_{v}(t) + n_{o}(0) = n_{v}(t) + n(0) - n_{v}(0).$$
 (15)

The relation (15) can be adjusted, assuming ideal behaviour, to give

$$P(t) = \frac{\int_0^h \frac{P(0) - P_v(z, 0)}{T(z, 0)} dz + \int_0^h \frac{P_v(z, t)}{T(z, t)} dz}{\int_0^h \frac{dz}{T(z, t)}}.$$
 (16)

COMPUTATION PROCEDURE

The system of equations (4)-(6) was solved numerically by the method of Katz and Ostermeier [12]. To solve the system of equations (10)-(12) and (14) we used the Crank-Nicholson implicit scheme. Due to the existence of the phase boundary, different network steps were used in different layers, and first derivatives at the interface were replaced by non-symmetrical difference formulas within the accuracy of the Crank-Nicholson layout. The corresponding systems of nonlinear equations have been solved by iterative quasilinearization [9]. The convergence of the iterative procedure was followed by comparing the temperature profiles resulting in single iteration steps. The calculation was terminated when the maximal temperature difference at an arbitrary point of the network was less than 0.01 K in two successive iterations.

EXPERIMENTAL EQUIPMENT AND METHODS

Temperature profiles in the layer of solid naphthalene in the system naphthalene-helium were used for the experimental verification of the calculated temperature of the subliming surface. A modified thermal diffusion cloud chamber was used for this purpose, as depicted in Fig. 2. The bottom and the lid of the chamber are formed from two aluminium discs 200 mm in diameter and 25 mm thick, the walls of a glass cylinder 33 mm high and 164 mm in diameter. Contact points between plates and the glass cylinder were sealed by Viton. The temperature of both plates is measured by jacketed NiCr-Ni thermocouples situated in probes

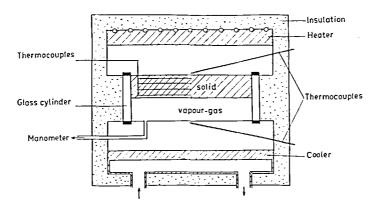


Fig. 2. Experimental apparatus.

bored so as to make the measurement points of the thermocouples end approximately in the centre of the plate and 0.5 mm below its surface. The temperature profile of the layer of naphthalene is measured by four NiCr-Ni thermocouples 0.15 mm in diameter, fixed in a holder. The holder body is made of Sklotextite, a material whose thermal diffusivity is similar to that of naphthalene, and has steel needles with eyes introduced parallel to the plate surface. The thermocouples are inserted to make them point radially 70 mm into the centre of the plate and end approximately above the thermocouple in the plate. The position of the thermocouples was controlled by a cathetometer (2.0, 4.2, 7.2 and 11.2 mm in the direction of the top plate). The thermocouples were calibrated in the entire region of temperatures using a standard Pt-100 resistance thermometer (Heraeus GmbH., Hanau/Main, F.R.G.). The bottom plate is kept at a constant temperature by contact with a cylindrical copper vessel, the top plate is heated programmatically by a heating body using a GP-1 graphical programmer and a TR-10 thyristor controller (Development Workshop of the Czechoslovak Academy, Prague). Temperatures are recorded with an EZ3 line recorder (Laboratory Instruments, Prague), and pressures in the chamber are measured by a mercury manometer. To prevent heat losses, the entire chamber is insulated with a thick layer of mineral wool.

Before starting actual measurements, we examined how the temperature of the plate surface corresponds to the temperature measured by the thermocouple in the probe at different heating rates. his was verified by comparing the melting point of benzophenone crystals and naphthalene, sprinkled on the plate surface in a position above the measurement point of the thermocouple. The temperature was recorded by the thermocouple itself. At a heating rate of 3 and 4 K min^{-1} the difference between both temperatures amounted to only $\pm 0.1 \text{ K}$, corresponding to the accuracy of temperature measurement. The equipment was further tested by measuring three temperature profiles at the steady state with the chamber filled by helium only. The temperature difference between the

plates in single runs was 14.4, 39.8 and 62.2 K at a helium pressure of 98 kPa. The agreement between temperature profiles calculated from plate temperatures and experimental values was within ± 0.2 K. These results prove the absence of free convection in the system (by the influence of colder walls) and verify the assumption that the thermal flux between the plates can be considered to be unidimensional for the given equipment geometry (large column diameter with respect to height). The results also indicate the accuracy of the determination of the position of the thermocouples and the separation of the plates, and the fact that the accuracy of measurement of the temperature by the thermocouples is not affected by thermal conduction in them.

The naphthalene layer (pure grade, sublimed, from Lachema, Brno), was prepared by melting the appropriate amount of crystalline naphthalene directly in the chamber. After its solidification and cooling to the laboratory temperature, the chamber was rinsed with helium gas (Messer Griesheim GmbH., Austria, 99.996% purity) from a pressure cylinder. The temperature of the bottom plate was kept constant during the measurement, while the temperature of the top plate was increased linearly with time. Both temperatures were recorded, as well as the temperature of the naphthalene layer and the total pressure in the chamber. Temperature profiles in the naphthalene layer were calculated from the temperatures of both plates and from the total pressure of the vapour-gas mixture.

The physical properties of the naphthalene and helium used are given in Table 1.

RESULTS AND DISCUSSION

Temperature profiles were measured after the described experimental method in the layer of solid naphthalene at different heating rates, thicknesses of the naphthalene layer and initial pressures in the chamber. To avoid convective mixing of the vapourgas mixture, the conditions of single measurements were selected so as to make the overall density of the

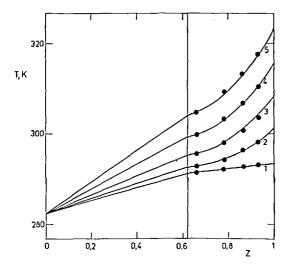
Table 1. Physical properties of naphthalene and helium

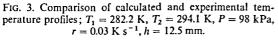
Naphthalene		-
M = 128.17	[g mol - 1]	
$\log P_{\rm eq} = 11.7059 - 2619.91/(T - 52.499)$	[Pa]	[13]
$C_{p,v} = -42.9422 + 0.6972T - 3.46 \times 10^{-4}T^2 + 2 \times 10^{-9}T^3$	[J K ⁻¹ mol ⁻¹]	[14]
$\hat{C}_{v,v}^{p,v} = -0.4 + 5.44 \times 10^{-3} T - 2.7 \times 10^{-6} T^2 + 1.56 \times 10^{-11} T^3$	[J K - 1 g - 1]	[14]
$C_{\rm p,s} = -0.346 + 5.507 \times 10^{-3} T$	$[J K^{-1} g^{-1}]$	[15]
$k_v = -3.98 \times 10^{-3} + 3.74 \times 10^{-5}T$	[W m ⁻¹ K ⁻¹]	[16]
$k_s = 0.7215 - 1.25 \times 10^{-3} T$	[W m ⁻¹ K ⁻¹]	[17]
$\rho = 1.375 - 4.64 \times 10^{-4} T$	[g cm ⁻³]	[18]
$\Delta H_{\text{subl}} = 72.5$	$[kJ mol^{-1}]$	[19]
Naphthalene-helium		
$\alpha = 1.0$		[10]
$D_{vg} = D_{vg}^0 R T^{s+1} / P$		
$D_{vg}^{00} = 1.424 \times 10^{-5}$	[mol s ⁻¹ m ⁻¹ K ^{-s}]	[20]
s = 0.75		[20]
$A_{vg} = 0.227, A_{gv} = 9.31$		[16]
Helium		
M = 4.003	[g mol ⁻¹]	
$k_{\rm g} = 3.72 \times 10^{-2} + 3.91 \times 10^{-4} T - 7.49 \times 10^{-8} T^2 + 1.29 \times 10^{-11} T^3$	[W m ⁻¹ K ⁻¹]	[21]
$C_{p,g} = 20.794$	[JK ⁻¹ mol ⁻¹]	[22]
$\hat{C}_{v,g} = 3.118$	[JK ⁻¹ g ⁻¹]	[22]

vapour-gas mixture highest at the bottom plate of the chamber and to let it decrease smoothly towards the naphthalene layer. The comparison of measured temperature profiles and calculated values is given for three particular cases in Figs. 3-5. Temperatures are plotted in single time intervals vs the dimensionless height Z = z/L. For the experimental run depicted in Fig. 3, the height of the naphthalene layer was 12.5 mm (Z = 0.62), the initial pressure in the chamber was 98 kPa and the rate of heating the top plate was 0.03 K s⁻¹. The profiles 1-5 correspond to times 0, 200, 470, 700 and 970 s. In Fig. 4, the conditions were as follows: thickness of naphthalene layer 18.8 mm (Z = 0.43);

initial pressure in the chamber 39.6 kPa; and heating rate 0.05 K s⁻¹. Temperature profiles 1–5 correspond to times 0, 240, 520, 960 and 1100 s, respectively. In the last case (Fig. 5), the thickness of the naphthalene layer was also 18.8 mm, the initial plate temperatures were 282.2 and 295.1 K, respectively, the initial pressure 98 kPa, and the rate of heating 0.02 K s⁻¹. Profiles 1–6 correspond to times 0, 500, 1040, 1550, 1960 and 2500 s, respectively.

The examples given indicate good agreement between the calculated and observed temperature profiles. The results thus verify the validity of the mathematical model used for the description of





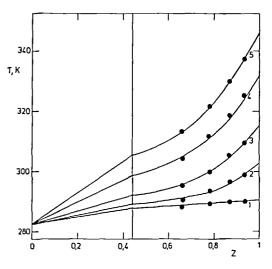


Fig. 4. Comparison of calculated and experimental temperature profiles; $T_1=282.4~\rm K, T_2=291.0~\rm K, P=39.5~\rm kPa,$ $r=0.05~\rm K~s^{-1}, h=18.8~\rm mm.$

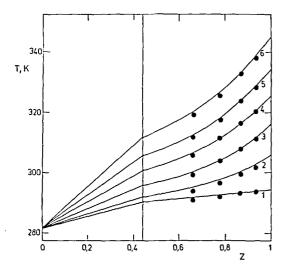


Fig 5. Comparison of calculated and measured temperature profiles; $T_1 = 282.2$ K, $T_2 = 295.1$ K, P = 98 kPa, r = 0.02 K s⁻¹, h = 18.8 mm.

transient heat and mass transfer in the respective equipment and the applicability of the mathematical solution. It is also validated that at the given system geometry there is no free convection in the system and that the heat flux between the plates can be well approximated by the unidimensional simplification. It follows also from the figures that the method applied enables a straightforward calculation of the temperature of the subliming surface. The practically linear temperature profiles in the vapour-gas mixture which were calculated indicate that the predominating mechanism of heat transfer in the experiments is convection, which substantiates the results of the earlier analysis [9]. From the point of view of computing temperature profiles, the entire problem can thus be simplified to heat transfer in the two-phase system, solid-inert gas.

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APPENDIX

The coefficients in equations (10)-(12) are defined as follows:

$$\begin{split} A &= k_{s}/\rho_{s}\hat{C}_{s}, \\ B &= (1/\rho_{s}\hat{C}_{s})(\partial k_{s}/\partial z), \\ C &= k_{vg}/\rho_{vg}\hat{C}_{v,vg}, \\ D &= (1/\rho_{vg}\hat{C}_{v,vg})(\partial k_{vg}/\partial z - v), \\ E &= -(1/\rho_{vg}\hat{C}_{v,vg})\{J_{v}(\partial \bar{H}_{v}/\partial z) + J_{g}(\partial \bar{H}_{g}/\partial z) \\ &\quad + RT\hat{c}(J_{v} + J_{g})/\partial z + (\alpha/P)\hat{c}[P_{v}(P - P_{v})(v_{v} - v_{g})]/\partial z \\ &\quad + P(\hat{c}v/\partial z)\}, \\ F &= RD_{vg}^{0}T^{s+1}/(P - P_{v}), \\ G &= RD_{vg}^{0}T\hat{c}[T^{s}/(P - P_{v})]/\hat{c}z, \\ H &= (\alpha RD_{vg}^{0}T/P)P_{v}T^{s-1}(\partial^{2}T/\partial z^{2}) \\ &\quad + \hat{c}(P_{v}T^{s-1})/\partial z(\partial T/\partial z) - RT\hat{c}[P_{v}N_{g}/(P - P_{v})]/\partial z \end{split}$$

 $+(P_v/T)(\partial T/\partial t)$

where the single variables are defined by the following relations

$$\begin{split} v_{\mathbf{v}} &= N_{\mathbf{v}}/c_{\mathbf{v}}, \quad v_{\mathbf{g}} = N_{\mathbf{g}}/c_{\mathbf{g}}, \\ v &= (\rho_{\mathbf{v}}v_{\mathbf{v}} + \rho_{\mathbf{g}}v_{\mathbf{g}})/\rho, \\ J_{\mathbf{v}} &= c_{\mathbf{v}}(v_{\mathbf{v}} - v), \quad J_{\mathbf{g}} = c_{\mathbf{g}}(v_{\mathbf{g}} - v), \\ \hat{C}_{\mathbf{v},\mathbf{v}\mathbf{g}} &= (\rho_{\mathbf{v}}\hat{C}_{\mathbf{v},\mathbf{v}} + \rho_{\mathbf{g}}\hat{C}_{\mathbf{v},\mathbf{g}})/\rho. \end{split}$$

The dependence of thermal diffusivity of the vapour-gas mixture on mixture composition is considered to have the

form

$$k_{vg} = x_v k_v / (x_v + A_{vg} x_g) + x_g k_g / (x_g + A_{gv} x_v)$$

and the partial molar enthalpies of the components

$$\vec{H}_{v} = \int_{T_{1}}^{T} C_{p,v} dT, \quad \vec{H}_{g} = \int_{T_{1}}^{T} C_{p,g} dT$$

where the temperature of the condensation surface T_1 is taken as the reference temperature. Some coefficients can be further adjusted; as given here, they were used for the difference transcription of equations (10)-(12).

TRANSFERT TRANSITOIRE DE CHALEUR ET DE MASSE DANS UN SYSTEME DIPHASIQUE SOLIDE-MELANGE VAPEUR ET GAZ AVEC SUBLIMATION

Résumé—On analyse le transfert transitoire de chaleur et de masse dans un système diphasique en sublimation solide-mélange de vapeur et de gaz. Le but est de déterminer la température de la surface du solide plan qui sublime dans un gaz inerte dans le cas où la température de la plaque chaussante est fonction du temps. La description mathématique du système est vérissée expérimentalement en déterminant les profils de température dans une plaque de naphtalène dans le système naphtalène-hélium. Un bon accord entre les profils de température calculés et mesurés justifie les approximations faites et vérisse les résultais d'une analyse antérieure du transfert variable de chaleur et de masse dans la dission thermique d'une chambre de Wilson.

INSTATIONÄRER WÄRME- UND STOFFÜBERGANG IN EINEM SUBLIMIERENDEN ZWEI-PHASEN-SYSTEM AUS FESTSTOFF UND GAS

Zusammenfassung—Der instationäre Wärme- und Stoffübergang in einem Zwei-Phasen-System aus Feststoffund Gas wird untersucht. Das Ziel der Arbeit ist die Bestimmung der Temperatur der sublimierenden Oberstäche eines Feststosses, der von einer Platte in eine Inertgasschicht verdampst wird, wobei die Plattentemperatur eine Funktion der Zeit ist. Die mathematische Beschreibung des Vorgangs wird experimentell bestätigt durch die Bestimmung des Temperaturverlaus in einer sesten Naphthalinschicht in dem System Naphthalin-Helium. Die gute Übereinstimmung zwischen den berechneten und gemessenen Temperaturverläusen rechtsertigt die angenommenen Vereinfachungen und bestätigt die Ergebnisse einer früheren Untersuchung des instationären Wärme- und Stossübergangs in der Thermodissusions-Nebelkammer.

ПЕРЕХОДНЫЕ ПРОЦЕССЫ ТЕПЛО-И МАССООБМЕНА В ДВУХФАЗНОЙ СИСТЕМЕ «СУБЛИМИРУЮЩЕЕСЯ ТВЕРДОЕ ТЕЛО – ПАРОГАЗОВАЯ СМЕСЬ»

Аннотация—Проведен анализ неустановившегося тепло-и массопереноса в двухфазной системе «сублимирующееся твердое тело – парогазовая смесь». Исследование предпринято с целью определения температуры сублимирующейся поверхности твердого вещества, испаряющегося с пластины в слой инертного газа в случае, когда температура пластины является функцией времени. Математическое описание системы проверено экспериментально путем определения профилей температуры в слое твердого нафталина в системе нафталин – гелий. Хорошее совпадение между рассчитанными и измеренными профилями температуры подтверждает справедливость используемых аппроксимаций и дает возможность проверить точность результатов анализа нестационарного тепло-и массопереноса, проведенного в термической диффузионно-конденсационной камере.