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HEAT AND MASS TRANSFER IN PHASE TRANSFORMATIONS

EVAPORATIVE COOLING OF BINARY DROPS IN A LOW-PRESSURE FLOW REACTOR

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A mathematical model of the evaporative cooling of binary drops in a flow reactor, accounting for the latent-heat liberation as a result of the phase transformation of the binary-drop components in the process of their condensation, the inverse processes of evaporation of these components, and the latent-heat liberation in the process of dissolution of one of the binary-drop components, has been developed. The numerical calculations were carried out for diluted ammonia spirits. It is shown that the evaporative cooling of binary drops in a flow reactor can be controlled by varying the compositions of the gas mixture and the solution and the ratio between their flow rates, and that the rate of cooling of drops can reach $2 \cdot 10^5$ *K/sec.*

Introduction. The evaporative cooling of small liquid drops with a radius of several microns at a low pressure in the gas phase is of importance for a number of modern technologies. In particular, this process can be used in ecologically clean air conditioning and ventilation systems. Moreover, the evaporative cooling of micron drops plays an important role in the production of supersaturated solutions used for obtaining nanoparticles by the promising method of pyrolysis of drops contained in solutions at a low pressure in a flow reactor [1]. In [2], experimental and theoretical data on the evaporative cooling of micron pure-water drops are presented. The mathematical model of the process being considered, developed in this work, can be used for the description, in the free-molecular approximation, of the heat and mass transfer between micron drops in the gas phase at a low pressure (lower than 100 Torr).

The bases of the kinetic description of the evaporation and condensation of gas drops were considered in [3]. Important additions to the kinetic description of the evaporation and condensation of vapor drops are presented in [4, 5]. Some problems on the evaporation of binary drops were considered in the diffusion approximation in [6].

The aim of the present work is to develop a mathematical model of the evaporative cooling of micron binary drops in a gas flow. This process was numerically simulated by the practically important example of micron drops of diluted ammonia spirits. The evaporative cooling of multicomponent drops of solutions opens up new possibilities for control of the formation of nanoparticles in the process of pyrolysis of drops at a low pressure (see [7]) with the use of multicomponent aqueous solutions containing ammonia.

Mathematical Model. We will consider the problem on the evaporative cooling of an ensemble of drops of an ammonia spirit in a flow aerosol reactor operating at a low pressure (Fig. 1). For simplicity, it will be assumed that, at the input of the reactor, the drops and the gas flow have equal temperatures and the molar fractions of ammonia in the liquid and gas phases are different. Nitrogen is used as the gas carrier.

It is also assumed that, at fairly low mass rates of the solution flow, the drops, on the average, are widely spaced — the distance between them is much larger than the diameter of a drop. Therefore, the process of heat and mass transfer occurring in the process of evaporation of an individual drop can be investigated without considering the action of the neighboring drops. We will consider a drop including N_{w} water molecules and N_{a} ammonia molecules at a temperature T_{d}. The radius of this drop R_{d} is defined as

$$\frac{4\pi R_{\mathrm{d}}^3}{3}\,\rho(x) = m_{\mathrm{a}}N_{\mathrm{a}} + m_{\mathrm{w}}N_{\mathrm{w}}\,, \tag{1}$$

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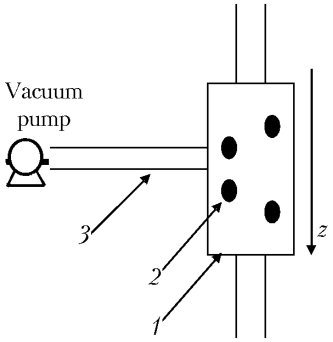

Fig. 1. Simplified scheme of a low-pressure aerosol reactor: 1) body of the aerosol reactor; 2) ammonia drops in water; 3) connecting tube.

where m_a and m_w are the ammonia and water molecular masses and $\rho(x)$ is the density of the ammonia spirit. The molar fraction of ammonia x is calculated in the following way:

$$x = \frac{N_a}{N_a + N_w},$$

the molar fraction of ammonia in the gas flow is determined analogously.

Since a drop moves uniformly with a velocity u equal to the velocity of the gas flow, the distance z travelled by the drop in the flow from the moment it enters the reactor (see Fig. 1) is used as the marching variable in our mathematical model. In the case where $dz = udt$, the equation defining the change in the molar fraction of ammonia in a drop moving in the reactor takes the form

$$\frac{dx}{dz} = \frac{1}{N}\left[(1 - x)\frac{dN_a}{dz} - x\frac{dN_w}{dz}\right], \tag{2}$$

where the number of molecules in the drop is determined as $N = N_a + N_w$. In the free-molecular evaporation regime, the change in the number of ammonia molecules in a drop is determined from the expression [3]

$$\frac{dN_a}{dz} = \frac{4\pi R_d^2}{u\sqrt{2\pi m_a k}}\left(\frac{p_{a,v}(T_m)}{\sqrt{T_m}} - \frac{p_{a,s}(T_d)}{\sqrt{T_d}}\right), \tag{3}$$

where $p_{a,v}$ is the partial pressure of ammonia in the gas phase, $p_{a,s}$ is the equilibrium pressure of the ammonia above the ammonia spirit, T_m and T_d are the temperatures of the gas mixture and the drop, and k is the Boltzmann constant.

The change in the number of water molecules in the drop N_w is determined from the equation

$$\frac{dN_w}{dz} = \frac{4\pi R_d^2}{u\sqrt{2\pi m_w k}}\left(\frac{p_{w,v}(T_m)}{\sqrt{T_m}} - \frac{p_{w,s}(T_d)}{\sqrt{T_d}}\right). \tag{4}$$

Here, $p_{w,v}$ is the partial pressure of water in the gas phase and $p_{w,s}$ is the saturation pressure of the water above the solution. Since $N_w \sim R^3$ at a constant molar fraction of ammonia, the left side of Eq. (4) is inversely related to the radius of a drop, i.e., the smaller the radius of the drop, the more intensive its mass exchange with the gas flow.

Below are two equations defining the changes in the numerical densities of molecules (of two types) in the gas flow (the rates of these processes are dependent on the total area of the interface of the drops, i.e., on the product of the number of monodisperse drops in a unit volume N_d and the area of the surface of a drop):

for the numerical density of the ammonia in the gas phase:

$$\frac{dN_{\mathrm{a,g}}}{dz} = -N_{\mathrm{d}} \frac{4\pi R_{\mathrm{d}}^2}{u\sqrt{2\pi k m_{\mathrm{a}}}} \left(\frac{p_{\mathrm{a,v}}(T_{\mathrm{m}})}{\sqrt{T_{\mathrm{m}}}} - \frac{p_{\mathrm{a,s}}(T_{\mathrm{d}})}{\sqrt{T_{\mathrm{d}}}} \right); \tag{5}$$

for the numerical density of the water vapor in the gas phase:

$$\frac{dN_{\mathrm{w,g}}}{dz} = -N_{\mathrm{d}} \frac{4\pi R_{\mathrm{d}}^2}{u\sqrt{2\pi k m_{\mathrm{w}}}} \left(\frac{p_{\mathrm{w,v}}(T_{\mathrm{m}})}{\sqrt{T_{\mathrm{m}}}} - \frac{p_{\mathrm{w,s}}(T_{\mathrm{d}})}{\sqrt{T_{\mathrm{d}}}} \right). \tag{6}$$

It is seen that the rate of change in the left sides of Eqs. (5) and (6) is directly proportional to the multiplier $N_{\mathrm{d}}R_{\mathrm{d}}^2/u$ and, therefore, is conveniently represented in the following form:

$$\frac{dN_{\mathrm{w,g}}}{dz} \sim \frac{Q_{\mathrm{w}}}{Q_{\mathrm{c}} R_{\mathrm{d}} u} \frac{\rho_{\mathrm{c}}}{\rho(x)},$$

where Q_{w} and Q_{c} are the mass rates of the water and gas flows through a unit area of the reactor cross section. In the case where the ratio between these flow rates remains unchanged, the mass exchange between the drops and the gas flow in the reactor intensifies with decrease in the radius of the drops.

The enthalpy of a binary drop $E(T_{\mathrm{d}}, x, N)$ is represented in the approximation

$$E(T_{\mathrm{d}}, x, N) = [m_{\mathrm{a}}N_{\mathrm{a}} + m_{\mathrm{w}}N_{\mathrm{w}}]\, c(x)\, T_{\mathrm{d}} - U_{\mathrm{a}}(x)\, N_{\mathrm{a}} - U_{\mathrm{w}}(x)\, N_{\mathrm{w}},$$

where U_{a} and U_{w} are the amounts of latent heat released by an ammonia molecule and a water molecule as a result of their phase transformation respectively. These amounts of heat depend on the molar fraction of ammonia. The enthalpy of a binary drop changes under the action of the gas-carrier, the water-vapor, and the ammonia-gas flows responsible for the energy transfer to a drop and from it. Representing these flows in the free-molecular approximation [3], we obtain the equation for the change in the temperature of the drop:

$$\frac{dT_{\mathrm{d}}(x)}{dz} = -\frac{1}{c(x)} \left\{ \begin{array}{l} -U_{\mathrm{dis}} \dfrac{dx}{dz} + \dfrac{3\,[\dot{N}_{\mathrm{a}}(m_{\mathrm{a}}c(x)\,T_{\mathrm{d}} + U_{\mathrm{a}}) + \dot{N}_{\mathrm{w}}(m_{\mathrm{w}}c(x)\,T_{\mathrm{d}} + U_{\mathrm{w}})]}{4\pi R_{\mathrm{d}}^3 \rho(x)} - \\ -\dfrac{3}{R_{\mathrm{d}}\rho(x)\, u} A \end{array} \right\}, \tag{7}$$

where the expression for A has the form $A = A_{\mathrm{g}} + A_{\mathrm{a}} + A_{\mathrm{w}}$. The energy fluxes for all the three components of the gas mixture can be represented, in turn, in the following form:

for the gas carrier:

$$A_{\mathrm{g}} = \frac{p_{\mathrm{g}}(T_{\mathrm{m}})}{\sqrt{2\pi k m_{\mathrm{g}}}} \left(\frac{c_{\mathrm{g}}T_{\mathrm{m}} + 0.5kT_{\mathrm{m}}}{\sqrt{T_{\mathrm{m}}}} - \frac{c_{\mathrm{g}}T_{\mathrm{d}} + 0.5kT_{\mathrm{d}}}{\sqrt{T_{\mathrm{d}}}} \right);$$

for ammonia:

$$A_{\mathrm{a}} = +\frac{1}{\sqrt{2\pi k m_{\mathrm{a}}}} \left(\frac{p_{\mathrm{a,v}}(T_{\mathrm{m}})\, T_{\mathrm{m}}}{\sqrt{T_{\mathrm{m}}}} (c_{\mathrm{a}} + 0.5k) - \frac{p_{\mathrm{a,s}}(T_{\mathrm{d}})\, T_{\mathrm{d}}}{\sqrt{T_{\mathrm{d}}}} (c_{\mathrm{a}} + 0.5k) \right);$$

for water vapor:

$$A_{\mathrm{w}} = +\frac{1}{\sqrt{2\pi k m_{\mathrm{w}}}} \left(\frac{p_{\mathrm{w,v}}(T_{\mathrm{m}})\, T_{\mathrm{m}}}{\sqrt{T_{\mathrm{m}}}} (c_{\mathrm{w}} + 0.5k) - \frac{p_{\mathrm{w,s}}(T_{\mathrm{d}})\, T_{\mathrm{d}}}{\sqrt{T_{\mathrm{d}}}} (c_{\mathrm{w}} + 0.5k) \right).$$

As follows from Eq. (7), the larger the radius of a drop, the smaller the influence exerted on its temperature by the energy transferred form the gas phase. Recall that the sum of the partial pressures in the reactor is assumed to be constant and is maintained with the use of a vacuum pump and a pressure transducer (Fig. 1) [1, 2, 7].

The heat of dissolution of an ammonia molecule in water U_{dis} is equal to

$$U_{\mathrm{dis}} = - T_{\mathrm{d}} \frac{dc\,(x)}{dx} ,$$

$c(x)$ is the heat capacity of the solution. In the calculations, the experimental value of the dissolution heat is used.

For determining the equilibrium partial pressure of the ammonia above its aqueous solution $p_{\mathrm{a,s}}$, Henry's law can be used [8, 9]: $x = \mathrm{H}p_{\mathrm{a,s}}$, where H is Henry's constant dependent on the temperature. For ammonia spirits, the values of H were experimentally determined in [8]. With the use of our mathematical model, we solve, in fact, the inverse problem: the value of $p_{\mathrm{a,s}}$ is determined by the known molar fraction of ammonia in a drop and its temperature. Thus, if the right side of Eq. (3) is larger than zero, the drop absorbs ammonia from the gas phase. As follows from the above-presented equations, we do not take into account the heat exchange between the walls of the reactor and the environment, because, first of all, it is assumed that the reactor walls are heat-insulating and the difference between the temperatures of the gas flow and the environment is fairly small.

Preparatory to the numerical calculations with the use of the model developed, we will perform qualitative estimates. For simplicity, a one-component water drop and a gas mixture consisting of water vapor and a gas carrier will be considered; the partial pressure of the water vapor will be assumed to be equal to p_{w}. Then, on the assumption that the flows of evaporating and condensing water molecules are equal, from (4) we obtain the equation for determining the minimum temperature of the pure-water drops T_{min}, attained in the process of evaporative cooling:

$$\frac{p_{\mathrm{s}}\,(T_{\mathrm{min}})}{p_{\mathrm{w}}} = \sqrt{\frac{T_{\mathrm{min}}}{T_0}} .$$

According to this formula, at a total pressure of 20 Torr in the reactor, an initial temperature of the solution of 300 K, and an initial pressure of 760 Torr, the partial pressure of the water vapor is determined as

$$p_{\mathrm{w}} = p_{\mathrm{w,s}}\,(300)\,\frac{20}{760} .$$

In this case, the minimum temperature of a drop $T_{\mathrm{min}} \approx 276$ K. In the case of evaporative cooling of a binary drop, the situation does not change radically, even though the formula becomes much more complex. As the numerical results presented in the figures below show, the value of T_{min} is not attained.

The experience on the study of the evaporative cooling of drops in a gas flow [10] suggests that a very important parameter is the ratio between the specific mass rates of the water and gas flows $Q_{\mathrm{w}}/Q_{\mathrm{c}}$. Moreover, in the evaporative cooling of binary drops, of importance is the composition of the drops and of the gas flow. In our case, of importance are the initial molar fractions of the ammonia in the drops and in the gas flow. The application of the dimensional theory [11] to our problem (the initial temperature and radius of the drops and the total pressure in the reactor are also considered as important parameters) gives the following results: for the radius of a drop found in the stationary state with the vapor-gas medium R_{st}, we obtain the relation

$$R_{\mathrm{st}} = R_0 F_1 \left(x, x_{\mathrm{g}}, \frac{Q_{\mathrm{w}}}{Q_{\mathrm{c}}} \right),$$

where R_0 is the initial radius of a drop and F_1 is a dimensionless function of the dimensionless arguments that, in turn, are similarity parameters of the problem on the evaporative cooling. The finite stationary temperature of a drop T_{st} is equal to

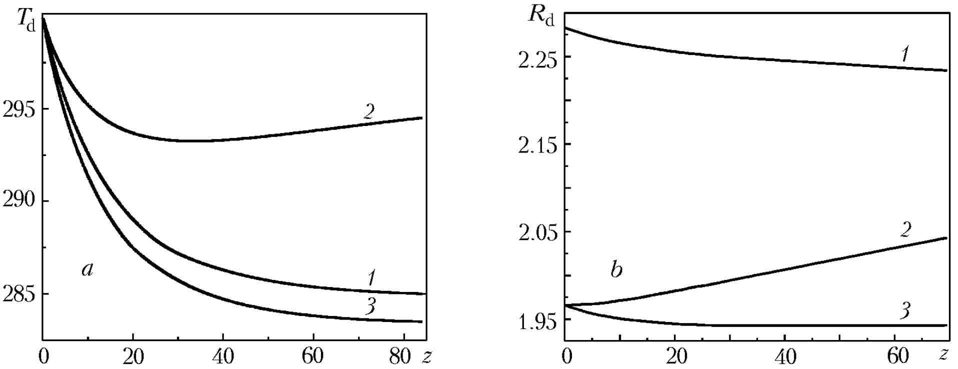

Fig. 2. Dependence of the temperature (a) and radius (b) of a drop on the distance travelled by it in the reactor: 1) x = 0.33, x_g = 0.05; 2) 0.05 and 0.26; 3) 0.05 and 0.05. Q_w/Q_c = 0.38, p = 60 Torr. T_d, K; R_d, z, μm.

$$T_{st} = T_0 F_2 \left(x, x_g, \frac{Q_w}{Q_c} \right).$$

It is notable that the total pressure in an aerosol reactor does not appear in the expressions for the finite radius and temperature of the drops since it mainly determines the dynamics of the evaporative cooling. The numerical results presented below support the conclusions of our qualitative analysis.

Results of Calculations. The system of equations (1)–(7) was solved numerically by the fourth-order Runge–Kutta method in the Mathcad-2000 medium. An important feature of our calculations is that the initial number of molecules in the water and the initial amount of ammonia in a drop and not the initial radius of the drop were prescribed. It is anticipated that the change to these variables will be useful in the theoretical investigation of the process of evaporation-condensation of multicomponent drops. The pressure of the saturated water vapor above an ammonia spirit was taken from the literature [12 and references there in].

Figure 2a shows the change in the temperature of approximately equal drops at different molar concentrations of ammonia in a drop and in a gas mixture. In this case, the total pressure in the reactor remained unchanged and equal to p = 60 Torr, and the initial temperature of the drops was 300 K. Of interest is first of all the high rate of cooling, equal to $\sim 2 \cdot 10^5$ K/sec, which is mush higher that the rate of evaporation of micron drops of a pure water [2]. Evidently, the reason for this is the larger values of the latent heat of the phase transition and the dissolution heat. The behavior of curve 2 shows that, after the initial stage of rapid evaporation of water molecules, the processes of absorption of ammonia drops from the gas phase become dominant. As a result of the liberation of the phase-transition latent heat and the dissolution heat, a micron drop begins to heat. This process will continue as long as the molar fractions of ammonia in the gas phase and in the drop are approximately equal (with regard for the difference between their temperatures). In this case, the rate of cooling of such a drop is lower by approximately an order of magnitude than in the cases represented by curves 1 and 3.

Figure 2b shows the change in the radius of the drop in the process of their evaporative cooling. It is seen that several variants are possible. If the molar fraction of ammonia in the gas phase is larger than that in a drop, the radius of the drop increases by several percent as a result of the evaporation of water molecules and the absorption of large ammonia molecules (curve 2). For curve 3 representing the case where the rate of cooling is largest, the radius of the drops changes by approximately 2%. It should be noted that this process differs substantially from the evaporation of pure-water drops in the aerosol reactor [2]. The change in the radius of a drop is largest in the case where water and ammonia evaporate simultaneously.

As our calculations have shown, the drops reach a stationary finite temperature as a result of the intensive mass exchange with the gas flow shortly after they enter the flow reactor. Figure 3a shows the dependence of the finite temperature of the drops on the total pressure in the aerosol reactor for different ratios between the mass rates of the water and gas flows Q_w/Q_c. The initial temperature of the drops was equal to 300 K. It is seen that, when the total pressure in the reactor increases, the finite temperature of the drops somewhat increases. The ratio between the

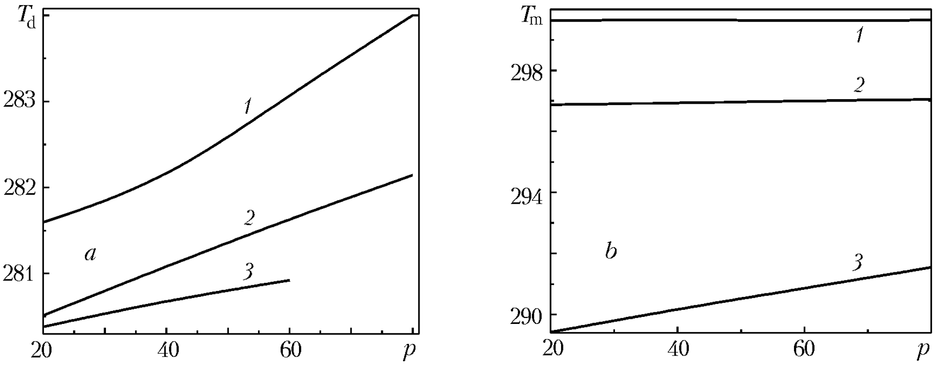

Fig. 3. Dependence of the finite temperature of a drop (a) and the gas mixture (b) on the pressure: Q_w/Q_c = 0.38 (1), 0.1 (2), and 0.011 (3); R_0 = 1.96 μm, $x = x_g = 0.05$. T, K; p, Torr.

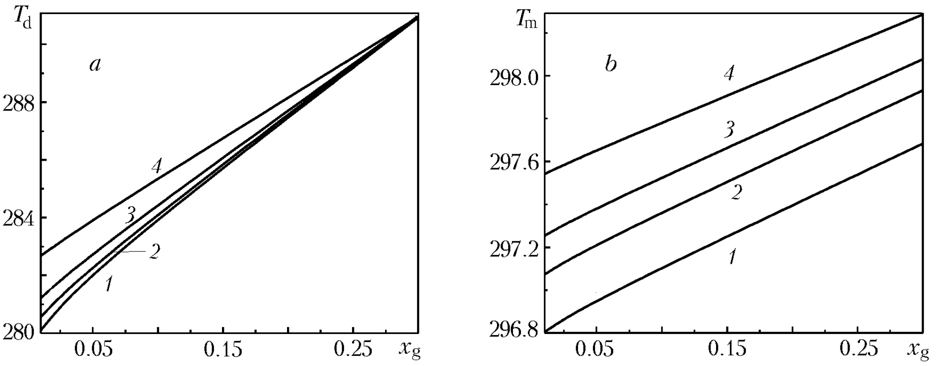

Fig. 4. Dependence of the finite temperature of a drop (a) and the gas mixture (b) on the molar fraction of ammonia in the gas mixture: R_d = 2 (1), 3 (2), 4 (3), and 6 μm (4). T, K.

mass rates of the water mixture and the gas flows influences the change in the finite temperature of the drops to a lesser extent than the change in the total pressure. The finite temperature of the drops reaches a minimum value at a minimum value of the parameter Q_w/Q_c on condition that the initial molar fractions of ammonia in the drops and in the gas phase are fairly close in value.

Figure 4a shows the influence of the initial molar fraction of ammonia in the gas flow on the evaporative cooling of the drops. It should be noted that the initial radius of a drop influences this process: the smaller the radius of a drop, the lower its temperature. For all the cases being considered, the initial molar fraction of ammonia in a drop was 0.05; the calculation was carried out for the case where the pressure in the aerosol reactor is constant and equal to 40 Torr and the ratio between the mass rates of the water and gas flows Q_w/Q_c = 0.1.

The mathematical model proposed also allows one to calculate the changes in the parameters of the gas phase in the reactor arising as a result of the evaporation of drops. Recall that the total pressure in the reactor is kept at a constant level with the use of a vacuum pump and a control system. Figure 3b shows the dependence of the finite temperature of the gas mixture in the reactor on the total pressure. The results are presented for R_d = 1.968 μm; the initial concentration of ammonia in a drop and in the gas mixture remains unchanged and equal to 0.05. It is seen from the graph that, at a small ratio between the mass rates of the water and gas flows Q_w/Q_c = 0.011, the temperature of the mixture remains practically unchanged. It is apparent that this property will be retained with further decrease in the ratio between the flow rates. However, the temperature of the gas mixture decreases with increase in the ratio between the flow rates to Q_w/Q_c = 0.1. In this case, the cooling of the drops is strongest at small values of Q_w/Q_c. Thus the value of Q_w/Q_c should be small for the pyrolysis of drops and large, at a fairly small pressure in a reactor, for ecologically clean air conditioning systems.

Figure 4 b shows the dependence of the finite temperature of the gas mixture on the initial molar fraction of ammonia in the gas flow. For all the cases, the initial molar fraction of ammonia in a drop is equal to 0.05.

The calculation data are in complete agreement with the above-presented analysis. It follows from these data that the finite temperature of the gas mixture depends weakly on the radius of the drops, as differentiated from the ratio between the mass rates of the two phases in the aerosol reactor and their initial temperatures.

Conclusions. A mathematical model of the evaporative cooling of a monodisperse ensemble of binary drops [11] with a radius of several microns in a low-pressure flow aerosol reactor has been developed, and numerical calculations of this evaporation have been carried out. All the calculations of the heat-and-mass transfer processes were performed in the free-molecular approximation. It was shown that the rate of cooling of binary drops can reach $\sim 2 \cdot 10^5$ K/sec. The process of evaporative cooling intensifies significantly when the molar fraction of ammonia in the drops is larger than the molar fraction of ammonia in the gas flow in the reactor.

In the reactor being considered, the cocurrent gas flow is cooled along with the drops. The degree of cooling of the gas flow depends mainly on the ratio between the mass rates of the drop flow and the gas flow. The larger the numerical density of the drops, the smaller the degree of evaporation of an individual drop required for it to reach the stationary state. Our calculations have shown that the heat and mass transfer in the reactor intensifies with decrease in the total pressure and the radius of the drops in it. An increase in the mass rate of the solution flow leads to a decrease in the intensity of the evaporative cooling of the drops. The present work is a continuation and a generalization of [2] for binary solutions.

NOTATION

$c(x)$, specific heat capacity of a liquid, J/(kg·K); c, heat capacity for one molecule, J/K; $E(T, x, N)$, enthalpy of a drop, J; F_1 and F_2, dimensionless functions of dimensionless arguments; H, Henry constant, Pa^{-1}; k, Boltzmann constant, J/K; m, mass of a molecule, kg; N, total number of molecules in a drop; N_a and N_w, number of ammonia and water molecules in a binary drop; N_d, number of monodisperse drops in a unit volume, m^{-3}; $p_\text{a,s}$ and $p_\text{w,s}$, saturation pressure of the ammonia and water above the solution respectively, Pa; $p_\text{a,v}$ and $p_\text{w,v}$, partial pressures of ammonia and water in the gas medium respectively, Pa; p_g, partial pressure of the gas carrier, Pa; Q, specific mass rate of flow, $\text{kg/(m}^2\cdot\text{sec)}$; R_d, radius of a drop, m; T, temperature, K; t, time, sec; u, velocity of travel of a drop, m/sec; U, latent heat of the phase transition of one molecule, J; U_dis, heat of dissolution of one ammonia molecule in water, J; x, molar fraction of ammonia; z, distance travelled by a drop, m; $\rho(x)$, density of an ammonia spirit, kg/m^3; ρ, density, kg/m^3. Subscripts: a, ammonia; c, gas flow; d, drop; dis, dissociation; g, gas-carrier; m, gas mixture; min, minimum; st, stationary state; s, saturated; v, in the vapor phase, w, water.

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