International Journal of Heat and Mass Transfer 52 (2009) 3842–3849

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Low pressure evaporative cooling of micron-sized droplets of solutions and its novel applications

Sergey P. Fisenko [*], Julia A. Khodyko

A.V. Luikov Heat and Mass Transfer Institute of National Academy of Sciences of Belarus, 15, P. Brovka St., 220728, Minsk, Belarus

ARTICLE INFO

Article history:
Available online 7 April 2009

Keywords:
Free molecular regime
Air cooler
Colloid solution
Nanoparticles growth

ABSTRACT

For free molecular regime the mathematical model of low pressure evaporative cooling of binary droplets in gas flow is developed. The model includes five ordinary differential equations and takes into account effects such as the release of the latent heat of condensation of both components and the release of the latent heat of dissolution. Simulations were made for weak aqueous solutions of ammonia. It was discovered that compositions of gas flow and the aqueous solution affect the rate of evaporative cooling of droplets. The ratio of mass flow of solution and gas flow is also an important parameter. The cooling rate of such binary droplets can reach the value of about 2×10^5 K/s.

As first applications we consider the air cooler based on evaporative cooling of droplets. For pressure of 20–80 Torr in aerosol reactor, it is shown that in the cooler with length of about 1 m temperature of air flow may drop to about 10–15 °C.

The second application is the formation of nanoparticle in evaporating multicomponent droplet with two volatile components. Simulation was made for aqueous solution of ammonia which is widely used by experimentalists and engineers now. Effects of the number of precursors in droplet and supersaturation in droplet on the final size of nanoparticles were investigated.

1. Introduction

Evaporative cooling of micron-sized droplets at a low pressure in a flow aerosol reactor can give new perspectives for a number of modern technologies. In particular, in this paper we will show that this phenomenon can be used for the creation of ecologically friendly ventilation and air conditioning systems. It is already known that the evaporative cooling of micron-sized droplets of solutions plays an important role in the novel method of nanoparticles production: a low pressure pyrolysis of droplets of solutions in a flow aerosol reactor [1]. There are no doubts that new applications of evaporative cooling of micron-sized droplets will be found soon! To mention only the application of it at the mass spectroscopy [2]!

Recently, experimental data and simulation results devoted to the evaporative cooling of micron-sized droplets of pure water have been published in [3]. In particular, it was discovered that the rate of cooling of pure water droplets reaches 2000 K/s at total pressure of 20 Torr in the flow aerosol reactor. For description of heat and mass transfer of evaporating droplets under low pressure we have to use the free molecular (kinetic) approach. Some fundamentals of the kinetic description of the evaporation–condensation of droplets are given in the famous classic book [4]. Recently, important applications of kinetic approach to evaporation have been presented in publications [5–6] and references therein. It is worth to mention that the evaporation of binary droplets was considered at the diffusion approximation in [7].

The evaporation of two and more volatile substances from droplets gives additional possibilities for the control of the cooling rate of droplets. For the control of the production of semiconductor nanoparticles in multicomponent droplets the evaporation of two volatile substances already has been used in the flow aerosol reactor [8].

Besides we consider that the evaporative cooling of micron-sized droplets could find application in coolers of gas flows [9]. High cooling rates of micron-sized droplets permit to design some compact heat exchangers. Cooling of stagnant air, based on the effect of evaporative cooling of water droplets, is widely used in greenhouses now.

The aim of this work is the development of the mathematical model of the evaporative cooling of micron-sized droplets with two volatile substances in gas flow. Additionally we give example of the calculation of performance of the air cooler based on a low pressure evaporative cooling of micron-sized droplets. We consider water and ammonia as two volatile substances. It is worth to emphasize again that developed mathematical model can be applied only for relatively low pressure in aerosol reactor; in other words at the free molecular approximation.

The paper is organized as follows. First we discuss the kinetics of evaporative cooling of binary droplets of a micron size in a flow

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[*] Corresponding author. Tel.: +375 17 284 22 22; fax: +375 17 292 25 13.
E-mail address: fsp@hmti.ac.by (S.P. Fisenko).

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doi:10.1016/j.ijheatmasstransfer.2009.02.017

Nomenclature

A	energy flow (W/m^2)
$c(x)$	specific heat capacity of solution (J/(kg K)
c	heat capacity per one molecule in gas phase (J/K)
D	diffusion coefficient (m^2/s)
E	droplet enthalpy (J)
H	the Henry constant (Pa^{-1})
I	source term
k	Boltzmann's constant (J/K)
L	characteristic length of cooler (m)
M	mass soluble impurity in droplet (kg)
m	mass of molecule (kg)
N	the number of something in droplet
P	pressure (Pa)
Q	mass flow rate per unit cross-section (kg/m^2 s)
R	radius (m)
r	radius of hole in the atomizer (m)
Re	the Reynolds number
S	supersaturation of solution in droplet
T	temperature (K)
t	time (s)
u	velocity (m/s)
U	heat per one molecule (J)
x	molar fraction of ammonia
z	distance (m)

Greeks

α	heat transfer coefficient of air flow (W/m^2 K)
λ	hear conductivity coefficient (W/m^2 K)
ρ	density (kg/m^3)
μ	dynamic viscosity (m^2/s)
σ	surface tension (J/m^2)

Subscripts

0	initial air in the clearance
1	aerosol reactor
2	clearance
A	ammonia
ag	ammonia in gas phase
as	saturated ammonia over aqueous solution of ammonia
av	ammonia vapor
c	colloid
cl	clearance
d	droplet
dis	dissociation
g	carrier gas
m	gas mixture
st	steady state
w	water
ws	saturated water vapor over solution
wv	water vapor

aerosol reactor. We apply kinetic approach based on the free molecular approximation because for low pressure the mean free paths of vapor molecules are significantly larger than droplet diameter. Next, we use our mathematical model of evaporative cooling for the description of the performance of an air cooler. In Section 4 we briefly discuss nanoparticles production in an evaporating multicomponent droplet. This process is greatly accelerated if there is a colloid solution in a droplet. Some preliminary results of this research have been published in [10] and reported in [11].

2. Mathematical model

The sketch of the flow aerosol reactor is shown in Fig. 1. The constant pressure is contained by vacuum pump and a controller with good accuracy. After the atomizer at the inlet of the aerosol reactor, the temperature of droplets is equal to that of the gas flow one. The composition of gas flow in the aerosol reactor is the same one as before aerosol reactor. The molar fraction of ammonia in droplets and one in gas flow may differ. We consider nitrogen as the carrier gas below.

For relatively low mass flow rates of solution the average distance between the droplets in the aerosol reactor is much larger than a droplet diameter. Therefore heat and mass transfer processes, related to evaporation, can be considered just for one droplet. Let us consider the droplet that has N_w of water molecules and N_a of ammonia molecules; to denote the droplet temperature as T_d. Then for radius R_d of this droplet we have algebraic equation

$$\frac{4\pi R_d^3}{3}\rho(x) = m_a N_a + m_w N_w, \tag{1}$$

where m_aиm_w are, respectively, molecular mass ammonia and water, $\rho(x)$ is the mass density of aqueous solution of ammonia. The molar fraction of ammonia x is determined by the following expression:

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

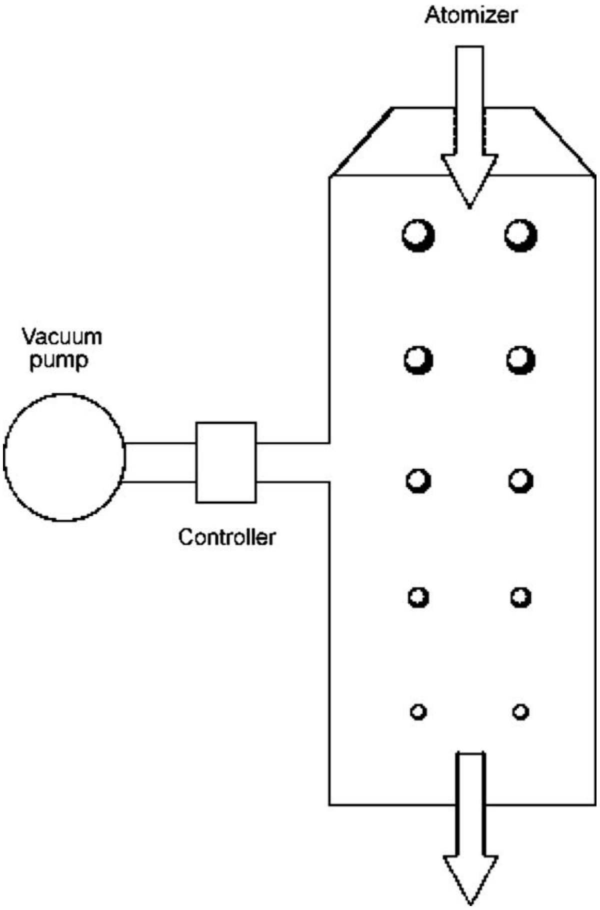

Fig. 1. Sketch of a low pressure aerosol reactor.

in a similar manner the molar fraction of ammonia is determined in the gas flow.

As droplets have the same velocity u as the gas flow in the reactor, in our mathematical model we select as the march variable the distance z from the inlet of the aerosol reactor (see Fig. 1). Based on the equality $dz = u\,dt$, the equation for description of evolution of molar fraction of ammonia in the moving droplet is

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

where the total number of molecules in the droplet N is equal to $N = N_a + N_w$.

For the free molecular regime the change in the number of ammonia molecules in the droplet N_a due to evaporation is described by the differential equation

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

where p_{av} is the partial pressure of ammonia in the gas phase, p_{as} is equilibrium pressure of ammonia over aqueous solution of ammonia, T_mиT$_d$ are the temperatures of gas mixture and droplet, k is Boltzmann's constant.

In a similar manner the change in the number of water molecules in the droplet N_w is described by the equation

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

where p_{wv} is the partial pressure of water vapor in gas phase, p_{ws} is the pressure of saturated water vapor over the aqueous solution.

The evolution of number densities of water and ammonia in the gas flow is described by two equations below. To emphasize that the rate of the mass exchange is dependent on the product of the number density of monodisperse droplets N_d and the area of the droplet surface $4\pi R_d^2$. For the change in the number density of ammonia in the gas flow we have equation

$$\frac{dN_{ag}}{dz} = -N_d\frac{4\pi R_d^2}{u\sqrt{2\pi k m_a}}\left(\frac{p_{av}(T_m)}{\sqrt{T_m}} - \frac{p_{as}(T_d)}{\sqrt{T_d}}\right), \tag{5}$$

for the number density of water vapor in gas flow we have equation

$$\frac{dN_{wg}}{dz} = -N_d\frac{4\pi R_d^2}{u\sqrt{2\pi k m_w}}\left(\frac{p_{wv}(T_m)}{\sqrt{T_m}} - \frac{p_{ws}(T_d)}{\sqrt{T_d}}\right). \tag{6}$$

It is worth to emphasize that the rates of the change in the left-hand of Eqs. (5) and (6) are directly proportional to the product $N_d R_d^2/u$. In particular, we can rewrite it in such manner

$$\frac{dN_{wg}}{dz} \sim \frac{Q_w}{Q_c R_d u}\frac{\rho_c}{\rho(x)},$$

where Q_w and Q_c are, respectively, the mass flow rate of water and gas per unit of the cross-section of the aerosol reactor. It is obvious that for given ratio of mass flows the intensity of mass exchange is inversely proportional to the droplet radius.

We write the enthalpy of binary droplet $E(T_d, x, N)$ in the following approximation:

$$E(T, x, N) = [m_a N_a + m_w N_w]c(x)T_d - U_a(x)N_a - U_w(x)N_w,$$

where U_a and U_w are respectively, the latent heat of phase transition per molecule of ammonia and water. The values of these parameters depend on the molar fraction of ammonia in the droplet. The change in the enthalpy of the binary droplet is determined by flows of carrier gas, water vapor and ammonia, transferring energy flows to the droplet and from it. In the free molecular approximation [4,5], we have the equation for the change in the droplet temperature:

$$\frac{dT_d(x)}{dz} = -\frac{1}{c(x)}\left\{-U_{dis}\frac{dx}{dz} + \frac{3[\dot{N}_a(m_a c(x)T_d + U_a) + \dot{N}_w(m_w c(x)T_d + U_w)]}{4\pi R_d^3\rho(x)} - \frac{3}{R_d\rho(x)u}A\right\}, \tag{7}$$

where for A the expression is

$$A = A_g + A_a + A_w.$$

In turn expressions of the energy flow transferring by every component are:for the carrier gas

$$A_g = \frac{p_c(T_m)}{\sqrt{2\pi k m_g}}\left(\frac{c_g T_m + 0.5kT_m}{\sqrt{T_m}} - \frac{c_g T_d + 0.5kT_d}{\sqrt{T_d}}\right),$$

for ammonia

$$A_a = +\frac{1}{\sqrt{2\pi k m_a}}\left(\frac{p_{av}(T_m)T_m}{\sqrt{T_m}}(c_a + 0.5k) - \frac{p_{as}(T_d)T_d}{\sqrt{T_d}}(c_a + 0.5k)\right),$$

for water vapor

$$A_w = +\frac{1}{\sqrt{2\pi k m_w}}\left(\frac{p_{wv}(T_m)T_m}{\sqrt{T_m}}(c_w + 0.5k) - \frac{p_{ws}(T_d)T_d}{\sqrt{T_d}}(c_w + 0.5k)\right).$$

To remind that total pressure in the aerosol reactor is maintained the constant and it's keeping by a vacuum pump and a controller (Fig. 1) [1,3,8].

The heat of ammonia dissolution in water U_{dis} per ammonia molecule we write as

$$U_{dis} = -T_d\frac{dc(x)}{dx},$$

where $c(x)$ is the heat capacity of aqueous solution of ammonia. For simulation we use experimental value of the dissolution heat.

For calculation of the equilibrium partial pressure of ammonia p_{as} over its aqueous solution we use Henry's law [12,13]:

$$x = Hp_{as},$$

where H is the Henry constant depending on temperature. For aqueous solution of ammonia value of the H constant had been experimentally determined in [13].

In this chapter we do not take into account the heat transfer between the wall of aerosol reactor and the environment. It will be the subject of our interest in Section 3 when we simulate an air cooler performance.

Before numerical simulation it is useful to make analytical qualitative estimations. For the sake of simplicity let us consider the pure water droplet and water vapor and carrier gas mixture. The partial pressure of water vapor is equal to P_w. Then for steady-state evaporation from Eq. (4) we obtain the equation for minimal temperature of pure water droplet T_{min}, which could be reached during evaporative cooling:

$$p_s(T_{min})/P_w = \sqrt{T_{min}/T_0}.$$

Thus for the total pressure in reactor 20 Torr and the initial temperature of solution 300 K, the partial pressure of vapor water is equal to

$$P_w = P_{ws}(300)\frac{20}{760}.$$

Finally, we have the value of the minimal temperature of pure water droplet: $T_{min} \approx 276$ K [3]. For evaporative cooling of binary droplet the physics of process is the same but formulas are more complicated ones. Nevertheless our simulation experience has shown that the minimal temperature of T_{min} is never reached (see Fig. 2), but it is an useful estimation of the cooling effect.

Also, it was established that the ratio of mass flow rates of water and gas Q_w/Q_c is a very important parameter [14]. For evaporative

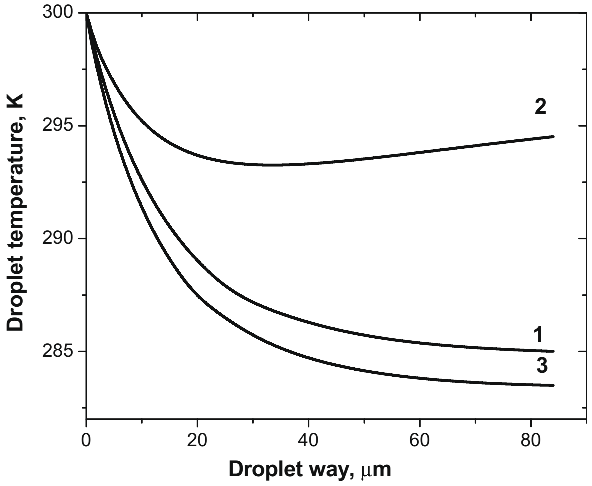

Fig. 2. Binary droplet temperature vs the way in low pressure aerosol reactor. For curve 1 $x = 0.33$ and $x_g = 0.05$, for 2 $x = 0.05$ and $x_g = 0.26$, curve 3 – $x = 0.05$ and $x_g = 0.05$; $Q_w/Q_c = 0.38$. $P = 60$ Torr.

cooling of binary droplets we, additionally, have two new parameters: initial molar fraction of ammonia in droplets and in gas flow.

2.1. Simulation results

The system of ordinary differential equations (3)–(7) has been numerically solved by the adaptive Runge–Kutta method of the forth order by means of Mathcad 2000 Professional. It was an important peculiarity of our simulation that we did not set the initial radius of the droplet. Instead we set the initial number of molecules of water and ammonia in the droplet. We do consider that the transition to these variables will be useful for theoretical investigation of evaporation–condensation multicomponent droplets also. It is worth to note that we use available data about the pressure of the saturated water vapor over the aqueous solution of ammonia [15] and references therein.

For different molar fractions of ammonia in the both phases the change in the droplet temperature is displayed in Fig. 2. For all variants the total pressure P was equal to 60 Torr, the initial temperature of droplets was equal to 300 K. First of all we would like to emphasize the most important result of our calculation in Fig. 2. The cooling rate is about 2×10^5 K/s. This value is significantly higher in comparison with evaporative cooling of micron-sized droplets of pure water [2]. It is obvious that the reason is the large value of the latent heat of evaporation of ammonia. Curve 2 gives the direct hint about it. Indeed we see that after fast initial evaporation of water molecules the adsorption of ammonia is the leading process; and the release of latent heat of the adsorption and the dissolution leads to heating of micron-sized droplet. The heating finishes when the molar fraction of ammonia in the gas phase and one in the droplet approximately equalize (taking into account the difference of their temperatures). For curve 2 the cooling rate is about the order smaller than one for curves 1 and 3.

The changes in the droplet radius on their way in the aerosol flow reactor are shown in Fig. 3. We see several scenarios. If the molar fraction of ammonia in gas flow is greater than that in the droplet then due to water evaporation and adsorption of larger ammonia molecules, the radius of droplet increases for several percents (curve 2). For the highest cooling rate (curve 3) the change in the droplet radius is about two percents. We see also that there is the substantial difference between current results and results obtained for pure water in [2]. Maximal change of droplet radius takes place when both components evaporate.

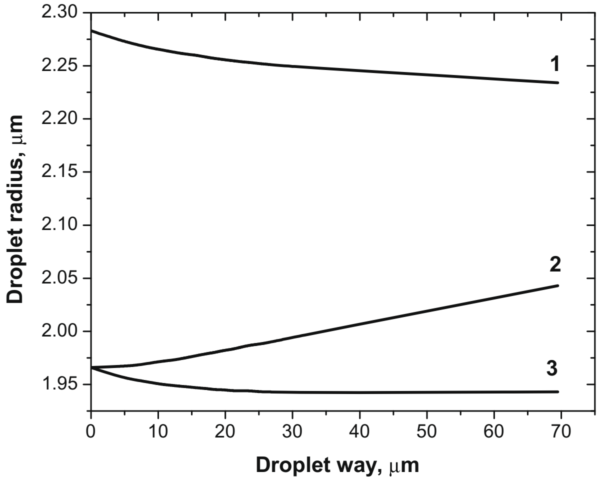

Fig. 3. Binary droplet radius vs its way in the low pressure aerosol reactor.

It was discovered that droplet temperature reaches the steady-state value very near inlet. The reason is the intensive mass transfer and release of latent heat of evaporation. For different ratios of flow rates of water и gas flow Q_w/Q_c the final temperature droplets versus the total pressure in the aerosol reactor is shown in Fig. 3. Initial temperature of droplets was 300 K. If we increase the total pressure we see that the final temperature of droplets also increases. The ratio of mass flow rates of solution and gas affects less significantly the value of the final temperature. Nevertheless minimal value of the final temperature is obtained for minima value of Q_w/Q_c if the initial molar fractions of ammonia in solution and in gas phase are practically the same.

Significant effect of the value of the initial molar fraction of ammonia in gas phase on evaporative cooling droplets in aerosol reactor is displayed in Fig. 4. For all cases in Fig. 4 the initial molar fraction of ammonia in droplet is equal to 0.05; the total pressure is 40 Torr and the ratio of mass flow rates $Q_w/Q_c = 0.1$. It is worthy to note that the smaller the droplet radius the more intensive is the process of the evaporative cooling.

Our mathematical model also permits to calculate the change in the parameters of gas flow in the reactor during the evaporation of droplets. The final temperature of gas flow in the reactor versus the

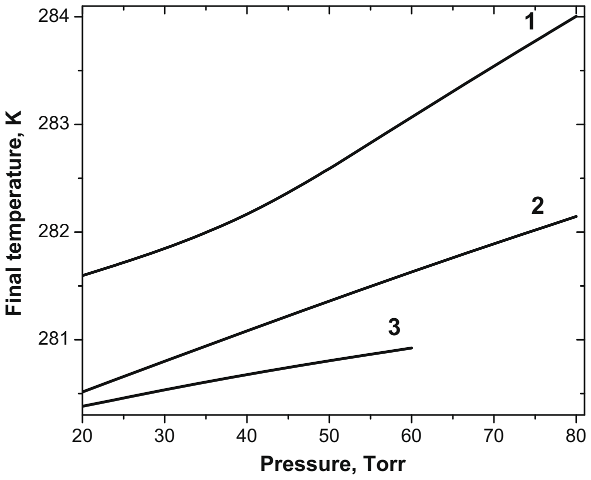

Fig. 4. The final temperature of droplets versus the total pressure in aerosol reactor. Curve 1 is for $Q_w/Q_c = 0.38$, curve 2 is for $Q_w/Q_c = 0.1$, curve 3 is for $Q_w/Q_c = 0.011$, the initial radius $R_0 = 1.96$ μm, $x = x_g = 0.05$.

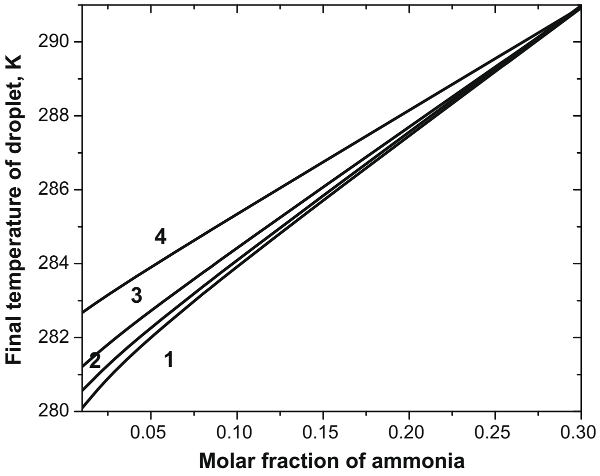

Fig. 5. The final temperature of droplets vs initial molar fraction of ammonia in gas mixture. Curve 1 is for the droplet radius 2 µm, curve 2 is for the droplet radius 3 µm, curve 3 is for the droplet radius 4 µm, curve 4 is for the droplet radius 6 µm.

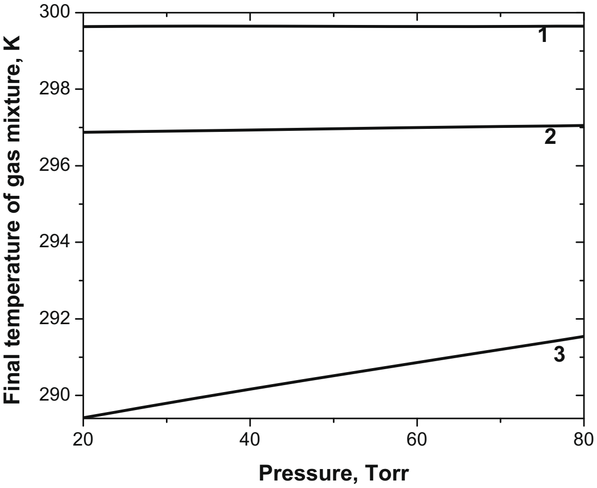

Fig. 6. Final temperature of gas flow vs the total pressure. Curve 1 is for $Q_w/Q_c = 0.011$, curve 2 is for $Q_w/Q_c = 0.1$, curve 3 is for $Q_w/Q_c = 0.38$.

total pressure is shown in Fig. 5. The simulation results are presented for initial droplet radius $R_d = 1.97$ µM; molar fractions of ammonia into droplets and in gas are equal to 0.05 and do not change during evaporative cooling. It is obvious that for small ratio of flow rates $Q_w/Q_c = 0.011$, gas flow temperature is practically constant. But for the case $Q_w/Q_c = 0.1$ the gas flow temperature decreases. It is interesting that droplet temperature most significantly drops for small ratio of flow rate Q_w/Q_c. Thus we can make conclusion that in nanotechnology applications one should use small ratio of flow rate Q_w/Q_c. We will show below that for successful performance of ecologically friendly air coolers we have to use low pressure for relatively large values of ratio of flow rates Q_w/Q_c (see Fig. 6).

3. Simulation of an air cooler

For calculation of parameters of air cooler let us consider it as two coaxial the flow-through cylinders (see Fig. 7). Also we consider that the external wall of the larger cylinder of the cooler is heat isolated one. The heated air flow is pumped in the clearance

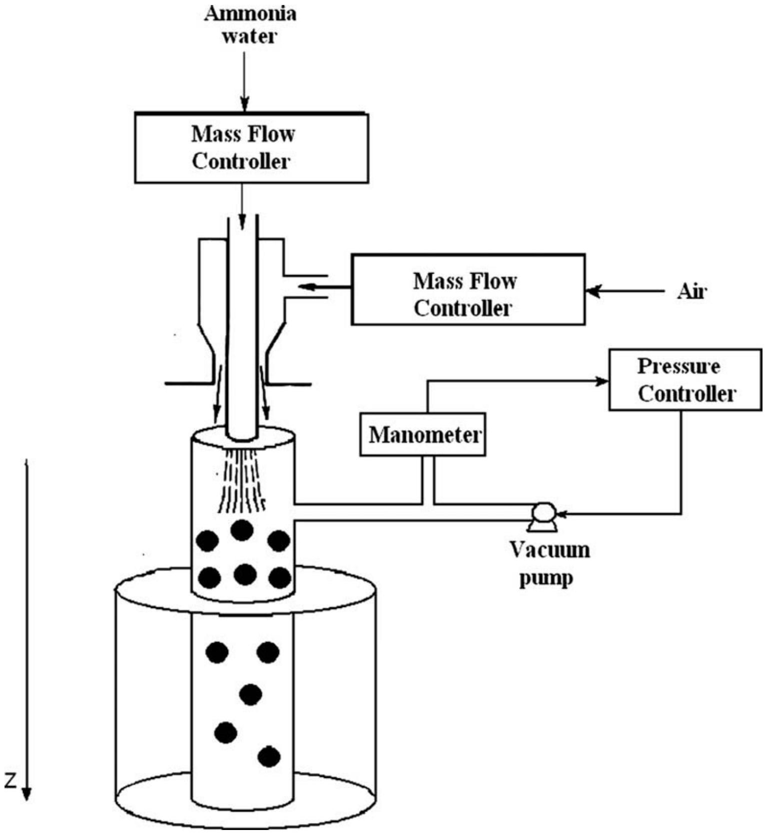

Fig. 7. The sketch of new air cooler.

between these two cylinders. Due to contact with cold wall of the internal cylinder the average temperature of this air flow drops. The gas flow with micron-sized droplets runs through the internal cylinder, which is a low pressure aerosol reactor and already considered above. Constant low pressure, which is at the range of 20–80 Torr in the internal cylinder, is maintained by vacuum pump with the pressure controller.

The pressure difference ΔP between pressure in the internal cylinder and atmospheric pressure is fitting in order to overcome the resistance of capillary forces in holes of atomizers. Indeed for liquid with the surface tension σ and hole with radius r we have the relationship

$$\Delta P \simeq 2\sigma/r.$$

It follows from this formula that the pressure difference between the atmospheric pressure and the pressure 20 Torr in the internal cylinder permits to obtain a micron-sized droplet of water at the room temperature.

3.1. Mathematical model of air cooler performance

Let us describe the fields of velocities and temperatures in the internal cylinder of the air cooler in one-dimensional approximation. We use the same approximation for clearance also. Then turbulent heat transfer coefficient $\alpha(Re)$ between air flow in the clearance and the surface of the internal cylinder is [16]

$$\alpha = \frac{0.025\,Re^{0.8}\lambda_g}{z},$$

where λ_g is the heat conductivity coefficient of air, z is the march variable; the Reynolds number Re is determined here as

$$Re = \frac{z\rho_g u_g}{\mu_g},$$

u_g is the velocity of air flow in the clearance, μ_g is the dynamic viscosity of air [3].

For one-dimensional approximation the equation for change in the the temperature of air flow in the clearance is

$$\frac{dT_2}{dz} = -\frac{2\alpha(T_2 - T_1(z))}{c_g\rho_0 u_g \pi(R_2^2 - R_1^2)}, \qquad (8)$$

where $T_1(z)$ is the averaged temperature of gas flow in the aerosol reactor on the same distance z from the inlet, $T_2(z)$ is the averaged temperature of gas flow in the clearance, ρ_0 is the density of air flow in the clearance, c_g is the heat capacity of air, R_2 and R_1 are radii of the external cylinder and the internal one.

The equation for change in the average temperature T_1 of the gas mixture in the internal cylinder is

$$\frac{dT_1}{dz} = \frac{2\alpha_1(T_1 - T_2)}{\pi R_1\rho_{1g}u_{1g}(c_w N_{wg} + c_a N_{ag} + c_g N_g)} + I, \qquad (9)$$

where c_w, c_a, c_g are, respectively, water vapor heat capacity, ammonia and gas-carrier per one molecule $\alpha_1 = 0.025\,Re_1^{0.8}\lambda_g/z$ is the heat transfer coefficient between the flow of gas mixture and the wall of aerosol reactor (internal cylinder) [16], Re_1 is the Reynolds number for the gas flow in the aerosol reactor; I is the term, describing the contribution of the evaporation of binary droplets into the cooling process:

$$I = \frac{2R^2 N_d A}{u_g(c_w N_{wg} + c_a N_{ag} + c_g N_g)}. \qquad (10)$$

It is worth to emphasize that droplets contribution in the heat transfer between droplets and gas flow is directly proportional to the interface area of all droplets. The source term I has more complicated dependence on the temperature difference between droplets and gas flow due to the free molecular regime, we denoted it already A.

For simulation of the air cooler initial conditions are: inlet temperature in the clearance and inlet temperature in the aerosol reactor, the total pressure into aerosol reactor, and the flow rate water and gas one in aerosol reactor. Also the mass flow rate of heated air through clearance is given. To remind that initial droplet radius is not given, we have only initial the number molecules of water and ammonia.

3.2. Qualitative estimations

As usually let us make qualitative estimation. For the constant heat transfer coefficient we can easily receive analytical expression for the temperature difference in the clearance after solving Eq. (8):

$$\Delta T(x) = \Delta T_0 \exp\left(-\frac{2\alpha x}{cQ}\right), \qquad (11)$$

where $\Delta T_0 = T_0 - T_{st}$ is the initial temperature difference of the gas flow in the clearance on the distance x. Then immediately we have the expression for characteristic length L of air cooler:

$$L = \frac{cQ}{2\alpha}, \qquad (12)$$

which is directly proportional to the flow rate of heated air and inversely proportional to the heat transfer coefficient. Any artificial enhancement of the value α has to decrease the characteristic length of the air cooler.

3.3. Simulation results

For simulation of the air cooler performance we numerically solve Eqs. (8)–(10) together with above-described equations of evaporative cooling (see Section 2). The most important simulation results are presented here.

The drop of the temperature of heated air in the clearance of the air cooler is shown in Fig. 8. We see that initial molar fraction of ammonia in droplets slightly affects the cooling rate of air flow in the clearance. We have discussed The physical fundamentals of this phenomenon already: the fast establishment of steady-state temperature of droplets.

For the air cooler with length two meters final temperatures of air flow are collected in Table 1 versus ratio of mass flow rates of water and carrier gas in the aerosol reactor. The total pressure

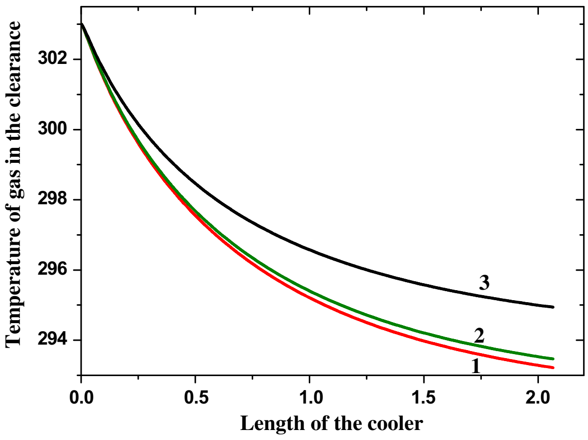

Fig. 8. Change in the temperature of air flow in the clearance. Curve 1 $\chi = 0.033$, curve 2 – $\chi = 0.05$, curve 3 – $\chi = 0.1$.

Table 1
Final temperature of air in the clearance vs parameters of the aerosol reactor.

$R_{d,\text{МКМ}}$	Final temperature (K)		
	$Q_w/Q_c = 0.38$	$Q_w/Q_c = 1$	$Q_w/Q_c = 3$
2	296.5	296.4	296.2
4	296.5	296.4	296.2
6	296.5	296.4	296.2
8	296.5	296.4	296.2
10	296.5	296.4	296.2

Table 2
Final temperature of air in the clearance vs the total pressure in aerosol reactor.

Pt (Torr)	Final temperature of air in the clearance (K)		
	$Q_w/Q_c = 0.38$	$Q_w/Q_c = 1$	$Q_w/Q_c = 3$
40	296.2	296.4	296.4
60	296.9	297	297
80	297.2	297.6	297.9

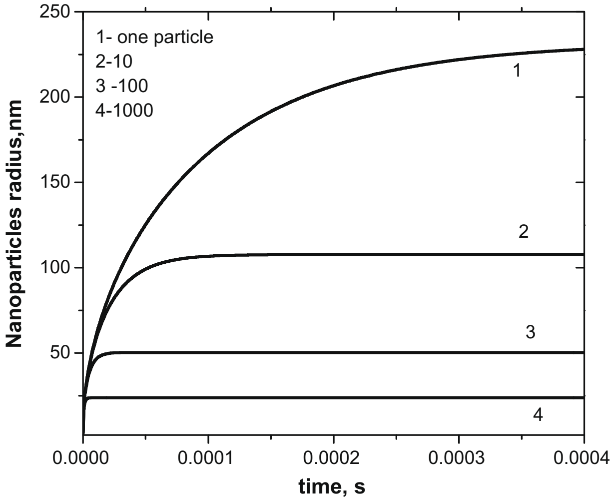

Fig. 9. Radius of nickel nanoparticles vs time.

was 40 Torr, the initial molar fraction of ammonia in droplet was equal to 0.1, we considered again pure nitrogen as carrier gas. We see that the performance of this air cooler is independent from varying many parameters of aerosol reactor.

For constant initial molar fraction of ammonia in droplets x = 0.1 and the initial droplet radius $R_d = 2$ µm effect of the total pressure in aerosol reactor on the final temperature of air in the clearance is presented in Table 2. We see that for the air cooler with length 2 m even the change in the total pressure slightly affects the efficiency of the air cooler performance. It is the manifestation of the different spatial scales of droplet cooling, discussed in Section 2, and the cooling of air flow in the clearance. For lower pressure we can make the air cooler with shorter length in order to observe the effect of total pressure.

Finally, let us illustrate the influence of such important dimensionless parameter of air cooler as the ratio of mass flow air in the clearance and one in aerosol reactor, Q_{cl}/Q_c. For total pressure of 40 Torr in aerosol reactor and the inlet temperature of air 303 K in the clearance our calculations showed that for $Q_{cl}/Q_c = 1$ the final temperature of air flow in the clearance falls at 8°C; for ratio $Q_{cl}/Q_c = 23$ final temperature drops only to 4 °C.

4. Growth nanoparticles in evaporating droplet

If we have a droplet of a multicomponent solution with several volatile components, we can expect that during evaporative cooling the formation of supersaturated solution takes place inside the droplets [1,8,11,17,19]. We determine the supersaturation of a component *S*:

$$S = X/X_{eq}, \tag{13}$$

where *x* and x_{eq} are, respectively, actual solubility of the component and equilibrium, depending only on the temperature. During the evaporation of volatile components the nominator increases in (13) if we consider nonvolatile substance at this range of temperature. For many substances the equilibrium solubility falls with decreasing temperature that enhances the effect of evaporative cooling.

For some cases we can expect even homogeneous nucleation in solution [11]. If there are some precursors in such droplets then there is a growth of nanoparticles in a supersaturated solution on precursors. The equation describing the diffusion growth of the nanoparticle in the droplet is

$$\frac{dR_n}{dt} = D\frac{\rho(t) - \rho_{eq}(T(t))}{R_n\rho_n}, \tag{14}$$

where *D* is the diffusion coefficient of soluble impurity, ρ_n is the density of nanoparticle, ρ and $\rho_{eq}(T(t))$ are, respectively, the average density of impurity in solution and the density of the equilibrium solution at the changing temperature *T*. The initial radius of nanoparticle is equal to the colloid particle. For beginning of the nanoparticle growth the radius of the precursor has to be larger than the Kelvin radius depending on the supersaturation of the solution and temperature [18]. Using (13) the difference $\rho(t) - \rho_{eq}(T(t))$ can be written as

$$\rho(t) - \rho_{eq}(T(t)) = \rho_{eq}(T(t))[S(t) - 1].$$

The average density of a soluble impurity inside evaporating droplet is determined by the conservation of impurity mass law:

$$\rho(t) = \frac{3}{4\pi R_d^3(t)}\left(M - N_c 4\pi R_n^3\rho/3\right), \tag{15}$$

where R_d is the droplet radius, *M* is the initial mass of soluble impurity, N_c is the number of colloid precursors in the droplet. It follows from the expression (15) that during droplet evaporation the average density of the impurity increases. Sometimes we have even the formation of supersaturated solution. Usually a solution supersaturation increases if the solution temperature falls. Simulation results of Eqs. (13)–(15) in conjunction with our equations of evaporative cooling (Section 2) are partially presented below.

We considered nickel as impurity because $Ni(NO_3)_2$ has been preliminary dissolved in the solution before the atomizer [19]. The initial droplet radius is 2 µm. In particular, the change in the radius of nickel nanoparticle versus time is shown in Fig. 9.

Unfortunately the ensemble of nanoparticles with diffusion interaction cannot grow with the same rate as it was suggested and displayed for Fig. 9. Often the Lifshitz–Slezov's instability leads to the formation of one large nanoparticle and a cloud smaller ones [18], such pictures of ensembles of nanoparticles are really observed in many experiments [19].

5. Conclusions

The mathematical model of evaporative cooling of binary droplets in low pressure flow aerosol reactor was developed. It was discovered that the best way for simulation is considering the change in the number molecules of different substances in the droplet. We took into account the release of latent heat of phase transitions and the heat of dissolution. All simulations have been made for weak aqueous solutions of ammonia at the free molecular approximation.

It was discovered that the cooling rate of binary droplets can reach value of about 2×10^5 K/s. If the molar fraction of ammonia in droplets is higher than one in gas flow the rate of cooling increases. Very fast cooled droplets start to chilly temperature of gas co-flow in aerosol reactor. Effect of cooling of gas flow depends on the total pressure in reactor and ratio of mass flow rates of droplets и gas flow.

We applied this model to the simulation of the performance of an ecologically friendly air cooler. The core of this air cooler is a flow low-pressure aerosol reactor. It was shown that for pressure of about 40 Torr in the aerosol reactor the heated air flow in the clearance lost about 10 °C on the way about one meter. Influence of mass flow rates and droplets radius was also investigated.

Also we considered the formation of nanoparticles in evaporating micron-sized droplets of multicomponent solutions with two volatile substances. Simulations have been made for widely used experimentalists mixture: aqueous solution of ammonia. It was shown that during evaporative cooling of droplets the formation of supersaturated solution relatively of one dissolved component takes place. If the supersaturation of the solution is high enough then there is a growth of a dissolved substance on the precursors. It is obvious that due to limited dissolved material in droplets the growth of nanoparticles is also limited, and its size depends on the number of precursors. We expect that the Brownian motion of growing nanoparticles and the evaporation rate of the droplet affect the formation of final structures from nanoparticles. This investigation is under progress now.

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