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Growth and evaporation of aerosol particles in the presence of adsorbable gases

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Abstract

Influence of adsorption of noncondensable molecules on the surface of phase transition on the rate of growth (evaporation) of aerosol particles (drops) is investigated theoretically. Combined manifestation of the effects that influence the rate of the growth of particles in the opposite directions is considered: decrease in surface tension on adsorption of a foreign gas and blocking of the surface of phase transition by the molecules adsorbed. The effect of the adsorption of the molecules of a noncondensable (in particular, buffer) gas on the rate of homogeneous nucleation and also on the growth of particles in chemical deposition is discussed.

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1. Introduction

Investigation of the mechanisms of formation and growth of particles by deposition from a gas phase is a highly urgent problem for both different kinds of atmospheric processes such as, for example, formation and growth of drops, and also of smog particles over industrial centres and processes in modern technology (in particular, manufacture of nanoparticles finding increasing application in engineering and medicine).

By the present time, a great number of both theoretical and experimental investigations has been carried out dealing with the problem of growth of particles from a gas phase. Review of these papers can be found, for example, in [1–4]. In most cases, the influence of a noncondensable component (in particular, of a buffer gas) is related to the resistance experienced by vapour molecules in transfer to a growing particle or from it (during its evaporation). In addition to the effect mentioned, molecules of a noncondensable component which are adsorbed on the phase transition surface can affect the growth (evaporation) of a drop by influencing phase transition.

Thus, in [5–7] investigation dealt with the influence of adsorption of the molecules of a buffer gas on the surface of clusters and also with the influence of a change in the pressure of saturated vapour on entering of foreign molecules into clusters on nucleation rate. The possibility of increasing nucleation rate with increase in the pressure of a buffer gas was related to a decrease in surface tension that leads to a decrease in the rate of cluster evaporation and hence to an increase in nucleation rate [5]. At the same time, the adsorbed molecules of the buffer gas can block the phase transition surface decreasing the rate of both evaporation and condensation. The present paper investigates combined manifestation of the mentioned effects in growth (evaporation) of aerosol particles (drops). In what follows, the effect of entering of the molecules of the noncondensable component into the particle (drop) which results in a change in the pressure of saturated vapour will be neglected for simplicity.

2. Influence of an adsorbable gas on the growth (evaporation) rate of a drop

It should be noted that the problem of the influence of an adsorbable gas on the growth and evaporation of drops can conditionally be divided into two parts. As mentioned above, the molecules of the buffer gas in

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Nomenclature

d	particle diameter
D	diffusivity of vapour molecules
I	density of resultant flux of vapour molecules into drop
k	Boltzmann constant
k_r	reaction rate constant
k_{r0}	preexponential factor in expression for reaction rate constant
Kn	Knudsen number
m	mass of a vapour molecule
m_a	mass of a molecule of adsorbable component
n	number concentration of vapour
n_a	number of adsorption sites per unit area
n_e	number concentration of saturation vapour above flat surface
n_∞	number concentration of vapour at infinity distance from the particle
P_a	pressure of adsorbable molecules of admixture (buffer) gas
Q_a	adsorption energy
Q_r	activation energy of chemical reaction
r	radial coordinate reckoned from particle centre
R	particle radius
S	n_∞/n_e
t	time
T	temperature
v	mean molecular velocity of vapour
v_p	particle growth rate
V_m	volume per molecule in particle

Greek symbols

α	evaporation (condensation) coefficient
γ	coefficient characterizing probability of heterogeneous chemical reaction in collision of molecule with surface
θ	surface coverage
λ	mean free path of vapour molecules
σ	surface tension

Subscripts

a	adsorbable gas
e	saturated vapour
m	molecule
p	particle
r	reaction
∞	infinite distance

which the drop is located, can adsorb on the drop (cluster) surface. As this takes place, an increase in the buffer gas pressure will lead to both an increase in the surface coverage of a drop by adsorbed molecules and a decrease in the diffusivity of vapour molecules in the buffer gas. On the other hand, the drop can be located in a nonadsorbable buffer gas in which, in addition to vapour molecules, there may be the molecules of a rather highly adsorbable component. The concentrations of both vapour and adsorbable component in a gas phase can be small enough, which enables one to consider the diffusivity of vapour molecules in the mixture of the buffer gas and an adsorbable component to be actually equal to the vapour diffusivity in the buffer gas (i.e., independent of the partial pressure of the adsorbable component). At the same time, at rather large adsorption times of the molecules of the adsorbable component, its presence in the buffer gas (even in sufficiently small concentrations) can exert a substantial influence on the processes occurring at the interface between the condensed and gaseous phases. Further we will consider this case.

It is supposed that the gas-particle system is isothermal and the condensation coefficient α for the vapour molecules incident on the particle surface which is free from the molecules adsorbed is independent of the drop size and equal to the evaporation coefficient.

It is known that an exact description of transfer processes in a gas phase in growth (evaporation) of aerosol particles of arbitrary sizes (and correspondingly of arbitrary Knudsen numbers) can be found only by solving the Boltzmann kinetic equation. However, the mathematical difficulties associated with the solution of the given equation lead to the necessity of obtaining rather simple expressions for mass and energy fluxes either on the basis of an approximate solution of the Boltzmann equation or with the use of simpler models. In particular, in the literature wide use is made of the "boundary sphere" model [1–4]. In this model, the entire flow region is divided into zones in which free molecular and continuous flow regimes are realized, respectively. The first zone is located between the particle surface and a certain spherical surface which is at a distance equal approximately to the free path of the gas molecules from the particle surface. It is assumed that beyond the abovementioned spherical surface the continuous flow regime of gas is realized, and mass transfer in the gas phase can be described by a diffusion equation. Matching is carried out on the boundary between these zones.

It should be noted that an exact value of the coefficient of proportionality δ between the distance at which the mentioned boundary sphere is located, and the free path of gas molecules is generally unknown and in different papers different values are used for this coefficient (including a zero value) [1,2]. At the same time, as noted in [3], if it is assumed that the diffusion equation describes the distribution of vapour concentration up to the particle surface, the expression obtained on its basis for the resultant flux of molecules into the particle gives a correct functional dependence at the Knudsen numbers much higher and much lower than unity. It should be remarked that zero value of the mentioned coefficient δ actually means the assumed validity of the diffusion equation up to the particle surface. The possibility of using the diffusion equation with assignment of a boundary condition on the surface of the particle whose radius is smaller than the free path of the vapour molecules is discussed in [8].

Without pausing to discuss numerous theoretical approaches available in the literature that enable one to obtain, with a certain degree of accuracy, expressions for the resultant flux of vapour molecules into the particle and taking into account the foregoing, in what follows we will use the diffusion equation to find a simple enough expression for the flux that gives limiting transitions for the free molecular and continuous flow regimes:

$$\frac{\partial n}{\partial t} = D\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial n}{\partial r}\right), \tag{1}$$

subject to the boundary conditions for $r = R$ and $r \to \infty$.

The problem of the growth (evaporation) of a drop will be considered in the quasi-stationary approximation. The stationary solution of Eq. (1) has the form

$$n = A - \frac{B}{r}, \tag{2}$$

where A and B are integration constants determined from the boundary conditions. When $r \to \infty$, we assume that $n = n_\infty$, whence it follows that

$$A = n_\infty. \tag{3}$$

To represent the second boundary condition, we will consider that the assumptions made in derivation of the Langmuir adsorption isotherm are satisfied. We suppose that both the adsorbable molecules of the noncondensable gas and the vapour molecules incident on the part of the surface occupied by the adsorbed molecules are elastically reflected. Here, we can write the following condition on the drop surface on the assumption of the Maxwell velocity distribution function for the incident molecules:

$$D\frac{\mathrm{d}n}{\mathrm{d}r}\bigg|_{r=R} = \alpha(1-\theta)\frac{v}{4}\left[n(R) - n_\mathrm{e}\exp\left\{\frac{2\sigma(\theta)V_\mathrm{m}}{RkT}\right\}\right], \tag{4}$$

where θ is the surface coverage by adsorbed molecules of the noncondensable gas, n_e is the number concentration of saturated vapour over the flat surface.

For the dependence of the surface tension on the pressure of the adsorbable gas P_a the following expression will be used [5]:

$$\sigma(P_\mathrm{a}) = \sigma(0) - n_\mathrm{a}kT\ln\left(\frac{P_\mathrm{a}+P_\mathrm{L}}{P_\mathrm{L}}\right), \tag{5}$$

where $\sigma(0)$ is the surface tension of the pure liquid. The value P_L can be represented in the form

$$P_\mathrm{L} = \frac{n_\mathrm{a}(2\pi m_\mathrm{a}kT)^{1/2}\exp\{-Q_\mathrm{a}/kT\}}{\beta\tau_0}, \tag{6}$$

where Q_a is the adsorption energy; τ_0 is the quantity characterizing the oscillation period of the adsorbed molecule in the direction perpendicular to the surface; m_a is the molecule mass of the adsorbable gas; β is the sticking coefficient of the molecule of the adsorbable gas for the part of the surface not occupied by the molecules adsorbed.

With Eqs. (2)–(5) and relation $\theta = P_\mathrm{a}/(P_\mathrm{a}+P_\mathrm{L})$ taken into account, we have the following expression for the density of the resultant flux of vapour molecules into the drop:

$$I = \frac{\alpha N\left[\dfrac{P_\mathrm{L}}{P_\mathrm{a}+P_\mathrm{L}} - \dfrac{1}{S}\exp\left\{\dfrac{2\sigma(0)V_\mathrm{m}}{RkT}\right\}\left(\dfrac{P_\mathrm{L}}{P_\mathrm{a}+P_\mathrm{L}}\right)^{G+1}\right]}{1+\alpha\dfrac{P_\mathrm{L}}{P_\mathrm{a}+P_\mathrm{L}}\dfrac{vR}{4D}}, \tag{7}$$

where $G = 2V_\mathrm{m}n_\mathrm{a}/R$, $N = n_\infty v/4$, $vR/4D = 3/(4Kn)$ and Knudsen number $Kn = \lambda/R$.

The growth rate of the particle v_p is defined as

$$v_\mathrm{p} = \frac{\mathrm{d}R}{\mathrm{d}t} = IV_\mathrm{m}. \tag{8}$$

As follows from Eq. (7) the critical (equilibrium) radius of the drop R^* depends on the pressure of the foreign gas. The value of R^* is given by the following equation:

$$R^* = \frac{2\sigma(0)V_\mathrm{m}}{kT[\ln S + G\ln(1+(P_\mathrm{a}/P_\mathrm{L}))]}. \tag{9}$$

It is shown in [9] that the drop growth rate as a function of P_a can be characterized by maximum for certain values of the parameters. The maximum owes its origin to the fact that the molecules of the buffer gas adsorbed at the drop surface exert a double influence on the growth of the drop. On the one hand, they can reduce the surface tension, which leads to a decrease in the rate of evaporation of molecules from a small drop and, correspondingly, to an increase in the rate of drop growth. On the other hand, the blocking effect of the adsorbed molecules results in the decrease in the rate of drop growth. The combined manifestation of the mentioned phenomena can lead to the indicated maximum.

Using expression (7), we will analyse the influence of size of the water drop and the dimensionless adsorbable gas pressure $P'_a = P_a/P_L$ on the resultant flux of the vapour molecules into a drop. We consider the second of the cases mentioned when the buffer gas is not adsorbed on the drop surface and the concentration of the adsorbable gas is rather small, which enables one to consider the vapour diffusivity a constant value (at a constant pressure of the buffer gas). For the evaporation and condensation coefficients of water the literature presents the values differing by orders of magnitude including the value equal to unity [10]. Further, we will use two values of α equal to 1 and 0.03 for a qualitative analysis of the α-dependence of a dimensionless flux of vapour molecules into a drop.

Fig. 1 presents the dependence of the dimensionless flux $I' = I/(\alpha N)$ on the water drop diameter d at 273 K for different values of S, α, and P_a/P_L. The value of n_a is taken to be equal to 6×10^{14} cm^{-2} [5]. The values of the surface tension of water and of the diffusivity of vapour molecules are supposed to be equal to 76.1 dyn/cm and 0.219 cm^2/s, respectively (the latter corresponds to the diffusivity of water vapour in air) [11]. It is seen from Fig. 1 that as the diameter of the drop increases, the influence of the adsorbed molecules on condensation (evaporation) decreases. When the drop grows ($I' > 1$), the function $I'(d)$ is characterized by a maximum, whereas when the drop evaporates ($I' < 1$), $I'(d)$ is the monotonic function. On fulfilment of the inequality $2\sigma V_m/RkT \ll 1$ and in the absence of the adsorbed molecules on the phase transition surface (which is reduced to the value P_a/P_L equal to zero), with Eq. (7) taken into account, we have the following expression for the value of the particle diameter $d_{\max}$ at which there is the maximum value of the resultant flux:

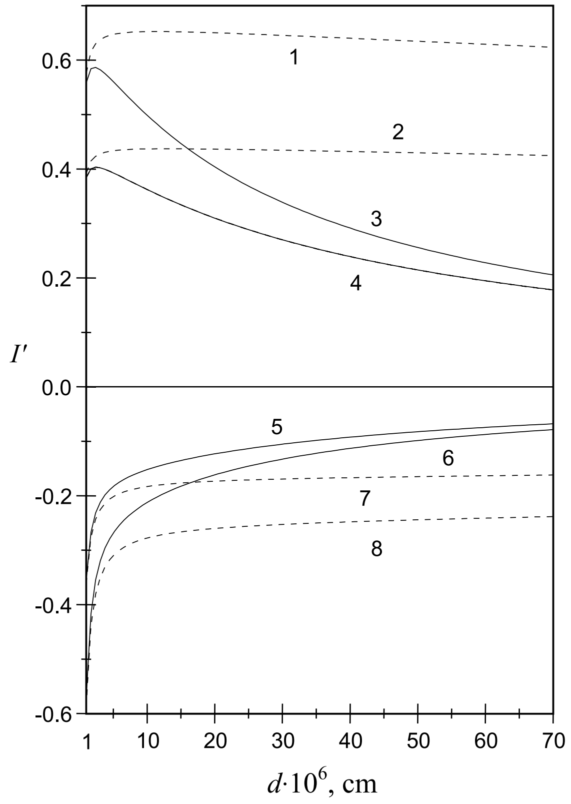

Fig. 1. Dependence of the dimensionless resultant flux of vapour molecules into a water drop $I' = I/(\alpha N)$ on the drop diameter at $T = 273$ K and $D = 0.219$ cm^2/s. Solid curves: $\alpha = 1$; dashed curves: $\alpha = 0.03$; 1, 3, 6, and 8: $P_a/P_L = 0$; 2, 4, 5, and 7: $P_a/P_L = 1/2$. The positive values of I' correspond to $S = 3$ and the negative ones to $S = 0.8$.

$$d_{\max} = \frac{\varphi\left[1 + (1 + (8D(S-1)/\varphi\alpha v))^{1/2}\right]}{S-1}, \tag{10}$$

where $\varphi = 4\sigma V_m/kT$. It follows from Eq. (10) that the maximum in the dependence of the resultant flux of vapour molecules into a particle on its diameter can be realized only for $S > 1$, with $d_{\max}$ being displaced to the region of larger diameters with a decreasing α.

The nonmonotonic dependence of the resultant flux of vapour molecules (and, correspondingly, of the rate of drop growth) on the drop diameter d is due to two factors affecting the growth rate in the opposite directions. For rather small drops, when a correction for the particle curvature for the saturated vapour pressure is substantial, an increase in the drop diameter leads to a decrease in the rate of molecule evaporation from the drop and, correspondingly, to an increase in the resultant flux of molecules into the drop. On the other hand, as the drop diameter increases, diffusive resistance to the supply of vapour molecules to the drop that is characterized by the second term in the denominator of expression (7) begins to increase, which leads to a decrease in the resultant flux of vapour molecules. These regularities can, in particular, influence the concentrations of the components in the particle during its condensation growth from a gas mixture [12].

Fig. 2 shows the dependence of the dimensionless resultant flux of vapour molecules into the water drop on the parameter P_a/P_L at different values of the drop diameter. It follows from Fig. 2 that with increase in the parameter P_a/P_L, the value of I' can both increase (this takes place at $d = 10^{-6}$ cm, which for $S = 1.27$ corresponds to the critical diameter of the drop) and decrease depending on the drop diameter. For rather large diameters of the drop, the I' value becomes actually independent of the presence of the adsorbed molecules of the foreign gas (curve 4 in Fig. 2). It is of interest to note that as follows from curves 4 and 5, the resultant flux of molecules into the drop can be the same for the drops of $d = 3 \times 10^{-4}$ and 10^{-6} cm at a certain value of P_a/P_L with all the remaining parameters being equal.

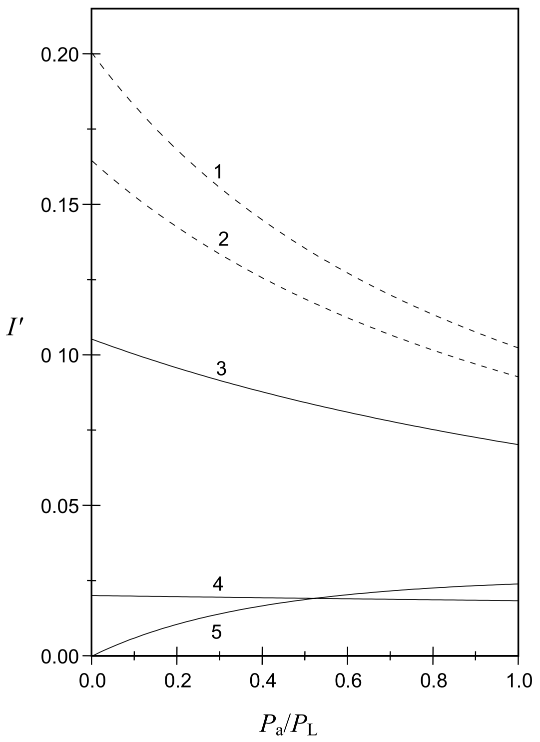

Fig. 2. Dependence of the dimensionless resultant flux of vapour molecules into a water drop I' on the parameter P_a/P_L at $T = 273$ K, $D = 0.219$ cm^2/s, and $S = 1.27$ with different values of the drop diameter and coefficient α. Solid curves: $\alpha = 1$, dashed curves: $\alpha = 0.03$; 1 and 3: $d = 3 \times 10^{-5}$ cm; 2 and 4: $d = 3 \times 10^{-4}$ cm; 5: $d = 10^{-6}$ cm.

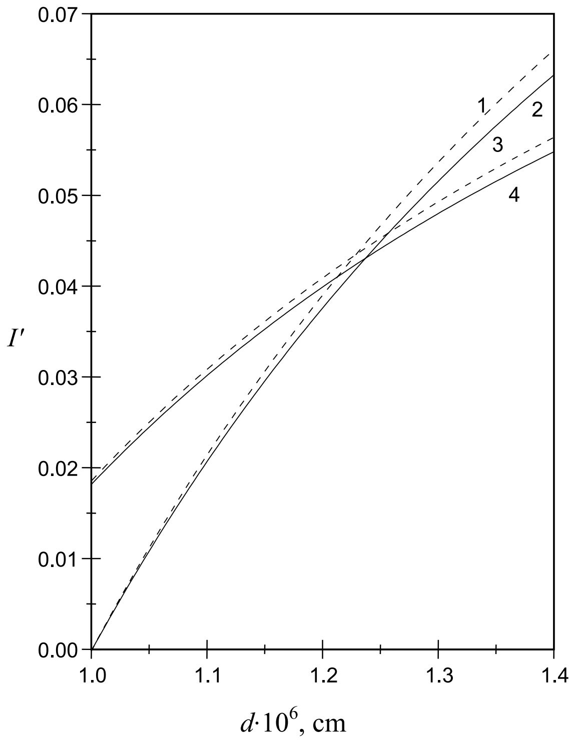

Fig. 3. Dependence of the dimensionless resultant flux of vapour molecules into a water drop I' on the drop diameter when the latter is close to the critical value for $T = 273$ K, $D = 0.219$ cm^2/s, and $S = 1.27$. Solid curves: $\alpha = 1$; dashed curves: $\alpha = 0.03$; 1 and 2: $P_a/P_L = 0$; 3 and 4: $P_a/P_L = 1/2$.

Fig. 3 presents the dependence of the dimensionless resultant flux of molecules into the water drop at small drop diameters which are close to the critical (equilibrium) one. It is seen from the curves given that the presence of the adsorbed molecules of the foreign gas can lead to a larger value of I' as compared to the value in the absence of these molecules. With the drop diameters used for calculations, the influence of the condensation coefficient on I' is insignificant.

3. Influence of the adsorption of the foreign (buffer) gas molecules on nucleation rate

It should be noted that adsorption of the molecules of a buffer gas on the surface of clusters can change the rate of homogeneous nucleation. Taking into account [1] and the fact that on the surface of clusters only a part of incident molecules can condense, namely, the part which, as in the previous case of deposition on a single drop, is assumed to be equal to $\alpha(1-\theta)$, we can write the following expression for the nucleation rate J:

$$J = \alpha(1-\theta)\left(\frac{2\sigma}{\pi m}\right)^{1/2}\frac{V_m n_1^2}{S}\exp\left\{-\frac{16\pi\sigma^3 V_m^2}{3(kT)^3(\ln S)^2}\right\}, \tag{11}$$

where n_1 is the number density of monomers, S is the saturation ratio.

A decrease in the surface tension due to the adsorption of buffer gas molecules on the surface of clusters leads to a change in the nucleation rate (to its increase) [5]. However, the presence of the factor $(1-\theta)$ in expression (11) can result in regularities analogous to those discussed above in the analysis of the process of deposition on a single drop. In the present case, there are also the factors that affect the nucleation rate in the opposite directions (blocking influence of the adsorbed molecules of the buffer gas leading to the decrease in the nucleation rate and the effect related to the decrease in the surface tension in the presence of these molecules that results in the increase in the nucleation rate). When the molecules of a noncondensable component enter into a cluster, the S value in Eq. (11) can also change [5].

4. Growth of particles by chemical deposition in the presence of an adsorbable foreign (buffer) gas

In the case of the growth of an aerosol particle during chemical deposition of molecules on the particle surface (such a process is often realized in the technique of manufacture of nanoparticles by deposition from a gas phase), molecule reevaporation from the growing particle is very weak and can be neglected in a number of cases ($S \to \infty$). Within the framework of assumptions made in derivation of Eq. (7) with consideration for Eq. (8), for the rate of particle growth we have

$$v_p = \frac{dR}{dt} = \frac{V_m \gamma N \dfrac{P_L}{P_a + P_L}}{1 + \gamma \dfrac{P_L}{P_a + P_L} \dfrac{v_r R}{4D_r}}, \qquad (12)$$

where the coefficient γ characterizes a fraction of the collisions of reactant molecules with the surface that lead to a heterogeneous chemical reaction, as a result of which a molecule (atom) is embedded into the particle, $N = n_{r\infty} v_r/4$, $n_{r\infty}$ is the number concentration of reactant molecules at infinity distance from the particle, v_r is the mean velocity of reactant molecules, D_r is the diffusivity of reactant molecules.

At $P_a/P_L = 0$ expression (12) reduces to the expression given in [13] for the rate of particle growth in chemical deposition. As follows from Eq. (12), for $Kn \gg 1$ the rate of the growth of a particle will decrease proportionally to the factor $1/(1 + P_a/P_L)$ with increase in the pressure of a foreign gas.

It should also be noted that in physical deposition the condensation (evaporation) coefficient usually has a weak dependence on temperature, whereas in chemical deposition the temperature dependence of the coefficient γ is quite appreciable. In the special case where the process of chemical deposition can be described by reaction of the first-order on the assumption of the Maxwell velocity distribution function of molecules incident on the surface, the coefficient γ in the one-stage approximation (without taking into account the occurrence of the molecule in an intermediate state of physical adsorption) may be presented as [14]

$$\gamma = \frac{4}{v_r} k_r = \frac{4}{v_r} k_{r0} \exp\{-Q_r/kT\}, \qquad (13)$$

where k_r is the reaction rate constant, Q_r is the activation energy of the chemical reaction.

It is seen from Eqs. (12) and (13) that in chemical deposition the rate of particle growth can increase with increasing temperature due to both rise of γ and decrease in P_L.

We can obtain an expression for the change in the particle radius with time from Eq. (12):

$$R(t) = F^{-1}\left\{\left[2FHt + (1 + FR_0)^2\right]^{1/2} - 1\right\}, \qquad (14)$$

where

$$F = \gamma \frac{v_r}{4D_r} \frac{P_L}{P_a + P_L}, \quad H = \gamma V_m N \frac{P_L}{P_a + P_L},$$

and R_0 is the particle radius at $t = 0$.

5. Conclusions

It follows from the foregoing that the presence of the adsorbed molecules of the buffer (admixture) gas on the surface of an aerosol particle (drop) affects both its growth and evaporation due to the blocking effect and the surface tension change. The molecules adsorbed can increase the rate of growth of a drop in the case of physical deposition (usual condensation) on the surface of a sufficiently small drop for the values of the S parameter which are close to the critical one. In chemical deposition, with neglect of reevaporation of the deposited molecules, an increase in the particle temperature can lead to an increase in the rate of particle growth due to both the increase in the reaction rate constant and the decrease in particle surface coverage by the adsorbed molecules of the buffer (admixture) gas. The considered effects related to the influence of the adsorbed molecules of the buffer gas on the growth and evaporation of particles (drops) should also be taken into account in investigation of processes of cluster origin and growth in the gas phase.

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