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DISTINCTIVE FEATURES OF PHASE TRANSITIONS IN THE FORMATION OF NANOSIZED AEROSOL PARTICLES

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The influence of a foreign gas on the formation of nanosized particles is investigated theoretically. The influence of heating the particles on the concentration of admixture molecules that are trapped in the process of condensation growth of the particles is considered.

Introduction. The formation of aerosol particles during their deposition from a gas phase is determined by processes like homogeneous nucleation, growth of formed clusters due to condensation of vapor molecules on their surface, as well as formation of aggregates in collision of clusters. The processes related to the formation and growth of nanosized aerosol particles usually occur in the presence of a buffer gas whose pressure greatly exceeds the pressure of the condensed component vapors. In this category we may place numerous phenomena occurring in both the atmosphere [1] and the technological processes connected with obtaining nanoparticles with definite properties [2]. When a basic component is deposited, molecules of a foreign (buffer) gas will also enter the condensed phase [3, 4], which in a number of case may substantially change its physicochemical properties. It should be noted that in the classical nucleation theory the parameters of a buffer gas do not enter explicitly into the expression that describes the nucleation rate. On the other hand, in a number of experimental works that investigate homogeneous nucleation the dependence of its rate on the buffer gas pressure was observed (a review of both theoretical and experimental works related to the influence of a buffer gas on the process of nucleation can be found, for example, in [5, 6]). Here, in most cases a decrease in the nucleation rate with increase in the pressure of the buffer gas was noted. However, there are also experimental data which reveal the inverse dependence — increase in the nucleation rate with the pressure of the buffer gas [5]. The authors of [5] relate this effect to the decrease in the surface-tension coefficient on adsorption of the buffer gas molecules on the surface of clusters, which leads to a reduction in the rate of molecule evaporation from the clusters. As noted in [7], the theoretical works on the influence of a buffer gas on the deposition process can be divided by convention into two groups: 1) works that, using thermodynamical, statistical, or kinetic approaches, study the effects resulting in deviation of the deposition rate from the value calculated on the basis of the classical nucleation theory, irrespective of a specific device in which the process considered takes place; 2) works devoted to the analysis of the influence of heat and mass transfer processes on deposition (nucleation) in specific devices (for example, in a thermal diffusion of continuous-flow chamber). Naturally, the transfer processes depend on the distribution of thermodynamic parameters in a concrete device, including the total pressure which is determined predominantly by the buffer gas pressure. It should be noted that in the works of the two groups mentioned not enough attention has been given to the kinetics of direct building of molecules into a growing particle. As noted in [7, 8], the presence of the adsorbed molecules of a buffer gas on the particle surface may both block incorporation of vapor molecules into the condensed phase and thereby decrease the rate of substance deposition and reduce the surface-tension coefficient, which leads to a rise in the growth rate of clusters. The total influence of the factors mentioned can result in the appearance of a maximum in the dependence of the cluster growth rate on the buffer gas pressure [7, 8]. Some problems of the influence of dimensional effects, external fields, and of the presence of a foreign adsorbable gas in the system on phase transitions on the surface of nanosized particles are considered in [9–11]. Moreover, an analysis was made

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of the change in the size of a critical cluster and correspondingly of the work of its formation under the effect of the factors noted. Below we consider the problems connected with the supply of vapor molecules to nanosized particles (clusters), evaporation of molecules from a cluster, as well as with trapping of the molecules of a foreign (buffer or admixture) gas by growing clusters.

Influence of a Buffer Gas on the Frequency of Collisions of Vapor Molecules with Nanosized Particles (Clusters). The expression for the nucleation rate in the framework of the classical nucleation theory can be written as (henceforth, for simplicity we will ignore the factor related to the nonisothermal effects that arise in nucleation) [12, 13]

$$J = k_Z \alpha_1 \nu_{1c} = k_Z \alpha_1 \frac{nv}{4} S_{cr} N_{cr} = k_Z \alpha_1 n \pi r_{cr}^2 v n \exp\left(-\frac{G}{kT}\right), \tag{1}$$

where $G = 4\pi\sigma_{cr}^2/3$.

It follows from (1) that the expression for the rate of homogeneous nucleation does not contain in an explicit form the dependence on the buffer gas pressure. This in particular is due to the fact that the flux density of the molecules incident on a cluster $nv/4$ is determined without regard for the resistance to the supply of vapor molecules to the cluster from a buffer gas. It should be noted that the coefficient of condensation of molecules on small particles (clusters) generally depends on their size [9, 14].

As noted in [15], the dependence of the frequency of collisions of vapor molecules with clusters on the concentration (pressure) of a buffer gas will manifest itself if for describing the vapor–cluster–buffer gas system the dusty gas model is used which treats the system mentioned as a gas mixture. In [16], based on this model, the transfer processes in highly porous bodies of globular structure with phase and chemical transformations on the walls of pores were considered. It should be noted that in [17] a mixture of molecules (monomers) and clusters made of these molecules are also treated as a gas phase. We will consider the frequency of collisions of vapor molecules with clusters using the dusty gas model. First we suppose that vapor molecules collide only with clusters. Here, for the frequency of collisions of vapor molecules with the critical clusters the following expression may be written:

$$\nu_{1c} = \frac{nv}{\lambda_{1c}}. \tag{2}$$

For the free path of a molecule (which is considered as a point) in the system of immovable spheres, with allowance for their volume and the possibility of mutual overlapping, we have [18]

$$\lambda_{1c} = \frac{1 - N_{cr} \frac{4}{3} \pi r_{cr}^3}{N_{cr} \pi r_{cr}^2 \left(1 - \frac{11}{16} N_{cr} \frac{4}{3} \pi r_{cr}^3\right)}, \tag{3}$$

where N_{cr} is the number density of the spheres (here, of the critical clusters).

With $N_{cr} \frac{4}{3} \pi r_{cr}^3 << 1$, for λ_{1c} we obtain

$$\lambda_{1c} = \frac{1}{N_{cr} \pi r_{cr}^2}. \tag{4}$$

Subject to (2), (4), we can show that the quantity nv/λ_{1c} coincides with the expression $n\pi r_{cr}^2 v n \exp\left(-\frac{G}{kT}\right)$ which enters into (1).

In the case where a buffer gas is present in a system, it is more difficult for a vapor molecule to "find" a cluster and to collide with it due to multiple collisions of this molecule with gas molecules. A coefficient which characterizes this effect should be introduced into expression (2). Assuming that the decrease in the number of collisions of vapor molecules with clusters, with allowance for the intermolecular collisions, is proportional to the ratio of the free path of a vapor molecule in the ternary vapor–buffer gas–clusters system λ_1 to the free path of a vapor molecule relative to the clusters λ_{1c}, for ν_{1c} we obtain

$$\nu_{1c} = \frac{nv}{\lambda_{1c}} \left(\frac{\lambda_1}{\lambda_{1c}} \right). \tag{5}$$

The quantity λ_1 is defined as

$$\frac{1}{\lambda_1} = \frac{1}{\lambda_{11}} + \frac{1}{\lambda_{12}} + \frac{1}{\lambda_{1c}} . \tag{6}$$

Since generally λ_{12} is significantly lower than both λ_{11} and λ_{1c}, we may consider that $\lambda_1 \approx \lambda_{12}$, i.e., $\dfrac{\lambda_1}{\lambda_{1c}} \approx \dfrac{\lambda_{12}}{\lambda_{1c}}$.

Thus, when relation (5) is used for ν_{1c}, the deposition rate decreases with increasing pressure of the buffer gas P_2, since $\lambda_{12} \sim 1/P_2$ (the buffer gas makes the collisions of condensing molecules with clusters difficult). It should be noted that in [19, 20] with the aid of the example of a single particle it was also shown that, when a substance is deposited from the gas phase onto a particle whose radius is much smaller than the free path of gas molecules, generally the resistance of the buffer gas will manifest itself. As mentioned above, the influence of the buffer gas in the pre-exponential term in (1) can reveal itself in the blocking effect of the molecules of the foreign gas that were adsorbed on the surface of phase transition [7]. Thus, the presence of the buffer gas in a system can decrease the rate of homogeneous nucleation due to both the occurrence of resistance to supply of vapor molecules to clusters and direct blocking of the surface of phase transition by the adsorbed molecules of the buffer gas.

Evaporation Energy and Condensation Coefficient for Nanosized Particles. It is known that the energy of evaporation of a molecule from a cluster depends on the cluster size. The drop model accounts for this fact with the aid of the Kelvin formula which reflects the influence of surface curvature on the saturation vapor pressure. Moreover, this formula contains the surface-tension coefficient. However, in the case of small enough clusters (especially dimers and trimers) it is desirable to have an expression for the evaporation energy that would not include the surface-tension coefficient, since for clusters consisting of several molecules it is difficult to interpret this quantity. In [9], an interpolation formula for the energy of molecule desorption from the surface of small clusters is presented. The formula accounts for the dependence of the desorption rate on the particle size and gives the limiting passages for desorption from a flat surface of massive material and for rupture of bonds between two different molecules (one of which is an adsorbent molecule and the other — an adsorbate molecule).

An analogous expression can also be obtained for evaporation of molecules from small particles (clusters). We consider a two-stage model of molecule evaporation, where a molecule initially passes from a volume onto a surface (into a self-adsorbed state) [21] and then evaporates (desorbs) from the surface. Here, for the evaporation energy E_e we may write

$$E_e = E_v + E_\infty , \tag{7}$$

where E_v is the energy of molecule passage from a volume onto a surface (in what follows its value is supposed for simplicity to be equal to the value of the given energy for a flat surface); E_∞ is the energy of molecule desorption from a flat surface of the same substance.

Then for a cluster of arbitrary size we can write an interpolation formula for the energy of evaporation of a molecule from the cluster. The formula gives the limiting passages for the energy of rupture of bonds between two identical molecules E_{11} and for the evaporation energy in the case of a flat surface:

$$E_e = E_v + E_\infty - (E_v + E_\infty - E_{11}) \left(\frac{d_m}{l} \right)^{\varphi} . \tag{8}$$

It follows from (8) that with $E_{11} < E_{\mathrm{v}} + E_{\infty}$ the energy of molecule evaporation from a small spherical particle will be lower than the energy of evaporation from a flat surface. It should be noted that generally the value of E_{v} in (8) depends on the particle size, which is due to the dimensional dependence of the probability of passage of a molecule from a volume phase onto a surface (i.e., from an absorbed state into an adsorbed one). The activation energy of passage of a molecule (atom) from the adsorbed state into the situation of absorption will also depend on the nanoparticle size, which will also influence the dependence of the probability of trapping (absorption) of admixture molecules by the particle on its size. Here, the decrease in the activation energy for both diffusion of molecules (atoms) in a nanoparticle and their passage from the adsorbed state to the absorbed one with decreasing particle size can be induced by the growth of the number of density vacancies in the particle.

Smaller values of the adsorption energy E_{a} usually correspond to lower sticking coefficients. For the sticking coefficient α we can write the expression [22]

$$\alpha = 1 - \exp\left\{-\frac{E_{\mathrm{s}}}{kT}\right\}, \tag{9}$$

where E_{s} is the specific energy of interaction of a gas molecule with a surface, which depends on the relationship between the masses of a gas molecule and an adsorbed atom, as well as on the energy of adsorptive interaction. For example, in the simplest case of the approximation which considers the collision of a molecule with a surface atom as elastic collision of free particles and with the fulfillment of the inequality $m_{\mathrm{g}}/M \ll 1$, where m_{g} is the mass of impacting gas molecule and M is the mass of an atom of the adsorbent lattice, for E_{s} we have [22]

$$E_{\mathrm{s}} \approx 4\frac{m_{\mathrm{g}}}{M}E_{\mathrm{a}}. \tag{10}$$

If the analogy between the adsorption–desorption and condensation–evaporation processes is taken into account, it is reasonable to expect that the condensation coefficient will also decrease with the evaporation energy. According to the above considerations, for a small particle this tendency can be related to the decrease in the effective energy of molecule evaporation from a particle on reduction of its size.

Formation of Nanosized Particles of Complex Composition. It should be noted that, when a basic component condenses, the molecules of the noncondensable component (buffer gas) will also be trapped by the condensed phase, which will lead to the formation of a condensed phase (including clusters and aerosol particles) of complex composition [3, 4].

Account for this effect can be realized on the basis of the model which was used in [3] for describing the trapping of admixture molecules in substance deposition from a gas phase. It should be mentioned that in [3] the admixture (relative to a condensed phase) molecules are considered to be those whose resulting flux into a growing particle is much smaller than the resulting flux of the molecules of the basic depositing component. Based on this determination, the molecules of a neutral buffer gas, whose heat of evaporation (desorption) from the surface of particles is much smaller than the heat of evaporation of the molecules of the basic depositing substance, can also be considered as admixture ones, despite the fact that their concentration in the gas phase is much higher than the concentration of the molecules of the basic component. The reason is the high rate of re-evaporation of the given molecules from the particle, as a result of which their resulting flux into the growing particle and correspondingly their concentration in the latter will be very small.

The resulting fluxes of molecules of a depositing component (I_1) and a buffer gas (I_2) into a growing particle subject to [3, 4] can be written as (further all the quantities related to the depositing component and the buffer gas will be marked by subscripts 1 and 2, respectively)

$$I_1 = \left(\alpha_{11}\frac{I_1}{I_1+I_2} + \alpha_{12}\frac{I_2}{I_1+I_2}\right)N_1 - \frac{I_1}{I_1+I_2}F_1, \tag{11}$$

$$I_2 = \left(\alpha_{21}\frac{I_1}{I_1+I_2} + \alpha_{22}\frac{I_2}{I_1+I_2}\right)N_2 - \frac{I_2}{I_1+I_2}F_2, \tag{12}$$

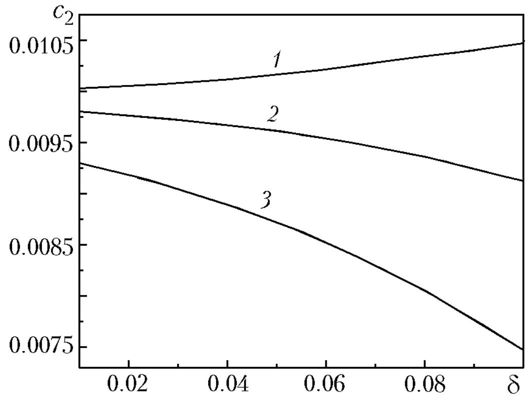

Fig. 1. Concentration of admixture molecules in a particle vs. the dimensionless temperature drop δ at $\frac{\alpha_2 N_2}{\alpha_1 N_1} = 0.01$, $\frac{F_{10}}{\alpha_1 N_1} = 0.01$, $\frac{E_1}{kT_0} = 19$: 1) $\frac{E_2}{kT_0} = 20$; 2) 18; 3) 17.

where α_{ij} is the coefficient of sticking of molecules of the ith component to the part of the surface occupied by molecules of the jth component; N_i is the flux density of molecules of the ith component ($i = 1, 2$) that are incident on the phase transition surface (which generally should be found with allowance for mass transfer in the gas phase). The quantities F_i which characterize the intensity of evaporation of components from the condensate take the form

$$F_i = n_s \left(\frac{kT}{2\pi m_i}\right)^{1/2} \exp\left\{-\frac{E_i(c_i, r)}{kT}\right\} = A_i \exp\left\{-\frac{E_i(c_i, r)}{kT}\right\}, \tag{13}$$

where the energy of evaporation of molecules of the ith component from the particle E_i depends generally on the concentration of components in the particle and its size.

After determination of I_1 and I_2 from (11), (12), we can find the concentration of components in the condensate. Next we suppose, for simplicity of the analysis, that $I_2/I_1 << 1$, E_i = const, $\alpha_{11} = \alpha_{12} = \alpha_1$, $\alpha_{21} = \alpha_{22} = \alpha_2$. Under the above-indicated assumptions, from Eqs. (11) and (12) the coefficient of trapping of admixture molecules β defined by the ratio of the density of the resulting flux of admixture molecules into the condensed phase to the flux density of the admixture molecules incident on the surface and the concentration of the admixture molecules in the condensate c_2 may be written as [3]

$$\beta = \frac{\alpha_2 I_1}{I_1 + F_2}, \tag{14}$$

$$c_2 = \frac{\alpha_2 N_2}{I_1 + F_2}. \tag{15}$$

An analysis of expressions (14) and (15) shows that, as the basic condensing component is deposited ($I_1 \neq 0$) in the case $\alpha_2 \neq 0$, trapping of molecules of a foreign (buffer or admixture) gas is always realized. With increasing condensation rate (i.e., the value of I_1) and other parameters being constant, the value of β will increase and that of c_2 decrease. The reason is that the probability of trapping of admixture molecules by the condensate rises with the condensation rate; however, when I_1 increases, for each trapped admixture molecule there is an increasing number of molecules of the basic material, which results in a decrease in the value of c_2.

In a number of cases, nanosized aerosol particles are formed and grow in the field of electromagnetic radiation (for example, on homogeneous nucleation of vapor formed as a result of evaporation of the condensed phase under the action of laser radiation or on nucleation in the atmosphere in the field of solar radiation). Of interest is the composition of the formed particles on their radiation heating. Let the gas temperature be equal to T_0 and the particle temperature be $T_1 = T_0 + \Delta T$ (particle heating is assumed to be uniform), with $\Delta T << T_0$. Here, the expression for the concentration of admixture molecules in the particle with regard to the above assumptions can be written as

$$c_2 = \frac{\dfrac{\alpha_2 N_2}{\alpha_1 N_1}}{1 - \dfrac{F_{10}}{\alpha_1 N_1} \exp\left(\dfrac{E_1 \Delta T}{k T_0^2}\right) + \dfrac{F_{20}}{\alpha_1 N_1} \exp\left(\dfrac{E_2 \Delta T}{k T_0^2}\right)}, \tag{16}$$

where $F_{i0} = n_{\mathrm{s}} \left(\dfrac{kT_0}{2\pi m_i}\right)^{1/2} \exp\left\{-\dfrac{E_i}{kT_0}\right\} = A_i \exp\left\{-\dfrac{E_i}{kT_0}\right\}$, $i = 1, 2$.

Further we consider for simplicity that the difference in m_i is small, so that we can assume that $A_1 = A_2$. Figure 1 shows the dependence of c_2 on the dimensionless temperature drop $\delta = \Delta T/T_0$ at different values of E_1 and E_2. As follows from the results presented, the value c_2 can both increase and decrease with increasing δ depending on the relationship between E_1 and E_2.

The quantities E_i and α_i in the presented expressions for c_2 were supposed to be independent of the particle radius. Generally, such a dependence can take place for fairly small particles (clusters). This leads to the concentration of admixture molecules in a growing nanosized aerosol particle (cluster) being nonuniform over the radius, since both the rate of particle growth (which is determined by the quantity I_1) and the rate of re-evaporation of molecules of the components from the particle in the general case depend on its radius. If the particle is characterized by an electric charge, the quantities E_i and α_i can generally differ from their values for a neutral particle, which, as follows from (14) and (15), will lead to the dependence of the quantities c_2 and β on the presence of the charge.

The foregoing enables us to conclude that with vapor condensation in the medium of a buffer noncondensable gas, particles of complex composition are formed, i.e., practically a solution is formed. Moreover, in the case of homogeneous nucleation the free energy of formation of a critical cluster will change. As noted in [23], because of the entry of molecules of the buffer gas into the clusters generated during homogeneous nucleation the problem of homogeneous nucleation of a vapor in a buffer gas should, in principle, be considered as binary nucleation.

Conclusions. Thus, some characteristic features of phase transitions at the initial stage of formation of nanosized particles that is related to the process of homogeneous nucleation and growth of the clusters generated have been considered in the work. The use of the dusty gas model for the determination of the frequency of collisions of vapor molecules with clusters is shown to lead to an expression which includes the dependence of the nucleation rate on the buffer gas pressure. Some problems of trapping of molecules of a foreign (in particular, buffer) gas by growing nanosized particles (clusters) have been analyzed. The influence of heating of a particle on the concentration of admixture gas molecules in it has been considered.

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NOTATION

c_i, concentration of molecules of the ith component in the condensed phase; d_{m}, diameter of an evaporating molecule; E_i, energy of evaporation of a molecule of the ith component from the condensed phase; G, free energy of formation of a critical cluster; I_i, density of the resulting flux of molecules of the ith component into the condensed phase; k, Boltzmann constant; k_{Z}, Zel'dovich factor; l, distance from the particle center to the center of the molecule (of the same substance) which was adsorbed on the particle (in the limiting case of two molecules $l = d_{\mathrm{m}}$); m_i, mass of a molecule of the ith component; N_{cr}, number density of critical clusters; n, number density of vapor molecules (monomers); n_{s}, total number density of molecules in a particle supposed to be independent of the concentration of components in a particle; P_2, pressure of a buffer gas; r_{cr}, radius of a critical cluster; r, particle radius; S_{cr}, surface area of a critical cluster; T, temperature; v, thermal velocity of vapor molecules; V_{m}, volume per molecule in a particle; α_1, condensation (sticking) coefficient of vapor molecules; α_2, sticking coefficient of molecules of a foreign (buffer or admixture) gas; β, trapping coefficient; $\nu_{1\mathrm{c}}$, frequency of collisions of vapor molecules with the critical clusters in a unit volume; λ_{1c}, free path of vapor molecules relative to clusters; λ_{11}, free path of vapor molecules relative to the same molecules; λ_{12}, free path of vapor molecules relative to buffer gas molecules; σ, surface-tension coefficient; φ, empirical parameter. Subscripts: c, cluster; cr, critical; m, molecule; e, evaporation; v, volume; a, adsorption; s, specific; ∞, value for a flat surface.

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