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CRITICAL SIZE OF AEROSOL PARTICLES IN PHYSICAL AND CHEMICAL DEPOSITION

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Problems related to the critical size of aerosol particles, when the vapor condensation coefficient depends on the particle size, the influence of electromagnetic fields, as well as on the presence of adsorbable foreign gases in the system, are studied theoretically. The problem of the critical size of particles in chemical deposition is discussed.

Keywords: *critical size, aerosol particles, deposition.*

Introduction. At present the increasing attention of researchers is drawn to the problems of obtaining different nanoobjects (in particular, nanoparticles, nanotubes) and systems made of them. This is related to an expanding field of application of nanosized systems both in various technological processes (for example, in the production of microelectronics elements, deposition of thin films, manufacture of efficient catalysts) [1] and in medicine [2]. Among the basic methods of obtaining nanoparticles is substance deposition from the gas phase. The problems of phase and chemical transformations on the surface of nanoparticles are of great interest in the field of investigation of atmospheric phenomena and protection against harmful emissions by modern industrial plants (ecological problems), since the process of generation and growth of water drops and smog particles depends substantially on the processes of deposition and evaporation of molecules on the surface of nanosized aerosol particles [3]. Essential characteristic features in the formation of nanoparticles appear in the case where the particles have an electric charge. For example, the presence of charged particles in a gas phase facilitates realization of the process of homogeneous nucleation.

As is known, the physicochemical properties of many substances in the form of nanoparticles differ from the properties of these substances in a sufficiently large volume [1]. However, most of the works related to investigations of nanoparticles are devoted to the study of their electric and magnetic properties. This is due to the wide use of nanoparticles in microelectronics. At the same time, there are a rather great number of unsolved problems as to the formation and growth of nanoparticles and systems made from them. The process of formation of nanoparticles in deposition from the gas phase depends substantially on the interaction of gas molecules with the surface of the particles. Here, characteristic features appear that owe their origin to the size effect. Thus, for example, in [4, 5] it is shown that in describing phase transitions on the surface of nanoparticles, the assumption, frequently used for macroscopic systems, on the equality of the condensation coefficient to the evaporation one (or the cruder assumption that these coefficients are equal to unity) may turn out to be incorrect in calculating the resultant flux of vapor molecules into a particle. Generally the condensation coefficient depends on the particle size, which affects the rate of the homogeneous nucleation and growth of the clusters formed. In the classical theory of homogeneous nucleation, one of the basic parameters that determine the nucleation rate is the diameter of a critical cluster at which the equilibrium condition in the particle–vapor system is realized. Small particles (clusters) can arise in the gas phase only at sufficiently high values of vapor supersaturation. Here, particles with size smaller than the critical one will evaporate to total disappearance, whereas those whose size is greater than critical will grow due to vapor condensation. This work considers some problems related to the determination of the critical (equilibrium) size of particles in both physical and chemical deposition of substance from a gas phase.

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Critical Size of Aerosol Particles in Physical Deposition (Condensation). To determine the critical size (diameter) of neutral spherical particles, the classical Kelvin formula that relates the critical particle diameter d_{cr} to the saturation ratio S is usually used. The latter is defined by the ratio of the partial vapor pressure to the saturated vapor pressure at a given temperature for a flat surface [6]:

$$d_{\text{cr}} = \frac{4\sigma V_{\text{m}}}{kT \ln S}\,. \tag{1}$$

This formula does not explicitly include the parameters that describe the interaction of gas molecules with the particle surface (in particular, the condensation and evaporation coefficients).

We will consider the problem of the critical size of particles in physical deposition on the basis of a kinetic approach, where the condition of the drop–vapor equilibrium is obtained from the condition of equality between the densities of the fluxes of evaporating and condensing molecules. It should be noted that the influence of different factors on the energy of evaporation of molecules from a small particle can be expressed through some additive term to the energy of evaporation from a flat surface [7]. Here, on the assumption that the velocity distribution functions for both impinging and evaporating molecules are Maxwellian ones, the expression for the resultant flux of molecules into the particle in the isothermal case can be written as

$$J = (1-\theta)\,\frac{v}{4}\left[\alpha_{\text{c}} n\,(R) - \alpha_{\text{e}} n_{\text{e}} \exp\left(\frac{Q}{kT}\right)\right], \tag{2}$$

where Q is the component of the energy of molecule evaporation from the particle due to such factors as its size (Q_1), electric field connected with the particle charge (Q_2), as well as external fields (in particular, resonance laser radiation) (Q_3):

$$Q = Q_1 + Q_2 + Q_3\,. \tag{3}$$

The following expression for Q_1 can be written with regard to the Kelvin effect:

$$Q_1 = \frac{2\sigma\,(\theta)\,V_{\text{m}}}{R}\,. \tag{4}$$

We note that generally the quantity Q_2 depends on the electrostatic and polarization components [7]. Further we suppose that at the particle center there is an elementary electric charge q. If only the electrostatic component is accounted for in Q_2, this quantity looks like (by analogy with [6], here the CGS electrostatic system of units is used)

$$Q_2 = -\frac{V_{\text{m}} q^2}{8\pi R^4}\left(\frac{1}{\varepsilon_{\text{g}}} - \frac{1}{\varepsilon_{\text{c}}}\right). \tag{5}$$

The quantity $1-\theta$ in (2) describes the blocking effect of the adsorbed molecules of a foreign gas relative to the evaporation and condensation processes; in the framework of the Langmuir adsorption theory it can be expressed in terms of the pressure of the foreign adsorbable gas P_{a} as

$$1-\theta = \frac{P_{\text{L}}}{P_{\text{a}} + P_{\text{L}}}\,, \tag{6}$$

where

$$P_{\text{L}} = \frac{n_0\,(2\pi m_{\text{a}} kT)^{1/2}}{\beta\tau}\,. \tag{7}$$

Subject to the equilibrium condition, when the resultant flux of vapor molecules into a particle is equal to zero, ignoring the flux of molecules due to the influence of a directed force on them (for example, with a drift of polar molecules in the field of the charged particle), the condition $n(R) = n_\infty$ is satisfied, where n_∞ is the number density of vapor molecules at infinity. The case where the particle charge changes the concentration of polar vapor molecules near the particle, leading to the nonequality $n(R) \neq n_\infty$ even in the absence of the resultant flux of vapor molecules into the particle, is considered in [8]. At $Q_3 = Q_2 = 0$ Eq. (2) yields the expression for the critical diameter of an uncharged particle:

$$d_{\mathrm{cr}} = \frac{4\sigma V_{\mathrm{m}}}{kT \ln (S\alpha_{\mathrm{c}}/\alpha_{\mathrm{e}})} . \tag{8}$$

Generally the condensation coefficient α_{c} for sufficiently small particles depends on their size [4]. This can be readily explained using as an example the generation of a dimer on collision of two vapor molecules and condensation of a vapor molecule on the surface of a macroscopic object from the same substance. It is known that the probability of realization of the first of the processes mentioned is much smaller than that of the second one. Thus, on an increase in the number of molecules in a formed particle (cluster) the probability of the sticking of molecules to it must increase. The sticking (condensation) coefficient can also be affected by electromagnetic fields, in particular, resonance radiation [9]. When a system is exposed to the resonance laser radiation, the condensation coefficient can change due to the excitation of vapor molecules in the radiation field. Here, with the use of the two-level model for transition of vapor molecules from a ground state to an excited one the critical diameter of a particle can be written as [10]

$$d_{\mathrm{cr}} = \frac{4\sigma V_{\mathrm{m}}}{kT \ln \left[\dfrac{\alpha_1 - \gamma(\alpha_1 - \alpha_2)}{\alpha_{\mathrm{e}}} S \right]} . \tag{9}$$

Here, α_1 and α_2 are the condensation coefficients of nonexcited and excited molecules, respectively, which for simplicity are assumed to be independent of the particle size, and the quantity γ is determined as

$$\gamma = \frac{n_2}{n_1 + n_2} , \tag{10}$$

where n_1 and n_2 are respectively the number densities of nonexcited and excited molecules, which are found from the equations

$$\frac{dn_1}{dt} = \frac{I_\nu \sigma_\nu}{h\nu} (n_2 - n_1) + \frac{n_2}{\tau_\nu} , \tag{11}$$

$$\frac{dn_2}{dt} = \frac{I_\nu \sigma_\nu}{h\nu} (n_1 - n_2) - \frac{n_2}{\tau_\nu} . \tag{12}$$

In a stationary state for γ we have

$$\gamma = \frac{(I_\nu \sigma_\nu / h\nu)\, \tau_\nu}{1 + (2 I_\nu \sigma_\nu / h\nu)\, \tau_\nu} . \tag{13}$$

At a constant value of S the critical diameter increases with I_ν in the case of $\alpha_1 > \alpha_2$ and decreases when $\alpha_1 < \alpha_2$. It follows from Eq. (8) that the critical particle diameter can be realized if the following inequality is satisfied:

$$S > \frac{\alpha_{\mathrm{e}}}{\alpha_1 - \gamma(\alpha_1 - \alpha_2)} . \tag{14}$$

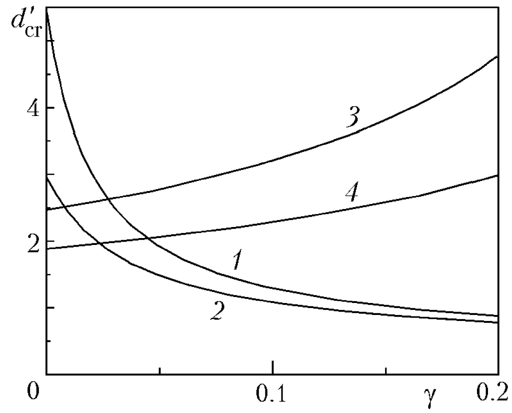

Fig. 1. Dependence of d'_{cr} on γ: 1) $\alpha_1 = \alpha_e = 0.1$, $\alpha_2 = 0.9$, and $S = 1.2$; 2) 0.1, 0.9, and 1.4; 3) 0.9, 0.1, and 1.5; 4) 0.9, 0.1, and 1.7.

The dependence of the dimensionless critical diameter $d'_{cr} = d_{cr}/(4\sigma V_m/(kT))$ on the quantity γ at different values of S for $\alpha_1 > \alpha_2$ and $\alpha_1 < \alpha_2$ is presented in Fig. 1. It is reasonable that generally a joint influence of thermal (particle heating) and resonance effects is realized. In particular, when the condensation coefficient decreases on excitation of vapor molecules, the thermal and resonance effects have the same tendency — they decrease the resultant flux of molecules into the particle. However, when the condensation coefficient of excited molecules is higher than that for nonexcited ones, the effects mentioned act in opposite directions, which leads to their partial mutual compensation.

On excitation of the molecules of a buffer gas in the field of resonance radiation, their sticking coefficient can be changed, which will result in a change in the concentration of adsorbed molecules of the buffer gas on the phase transition surface. In turn, this entails a change in the surface-tension coefficient and, correspondingly, in the critical particle size.

In the case of nanosized aerosol particle, the presence of an electric charge in it can change the condensation coefficient of polar vapor molecules. This is due to the following factor. Because of the sufficiently low value of the time of rotational relaxation, the polar molecules (dipoles) impinging on the charged particle can be oriented relative to the particle surface in a certain way depending on the charge sign. Incorporation of the molecules incident on the particle into the condensed phase may depend on the orientation of the given molecules relative to the surface of the condensed phase (i.e., on which of the dipole ends collides with the particles). For example, water molecules condense more easily on aerosol particles with a negative charge, when an electric field orients dipoles by their positive charge to the particle surface. To condense water vapor on a negatively charged particle, less vapor supersaturation is needed than in condensation on a particle with a positive charge [6]. This dependence can appear in the case of asymmetry in the interaction of dipole vapor molecules with particles having charges of different signs, which, in particular, can be due to the difference in the condensation coefficients for the polar molecules incident on the aerosol nanoparticles with charges of opposite signs.

In [11], the problem of the density of a resultant vapor flux into a charged nanosized particle is considered. It is seen that this quantity can be characterized by two extreme — minimum and maximum. This is explained by joint manifestation of the Kelvin effect, the influence of an electric charge, and by the dependence of vapor molecule supply to the particle on the Knudsen number.

Adsorption of molecules of a foreign gas on the surface of nanoparticles can also substantially change the rate of their formation [12]. This is connected with the blocking effect of adsorbed molecules relative to phase transitions on the surface of the particles, as well as with the change in the surface-tension coefficient due to the adsorption of the foreign gas molecules. As follows from the calculations carried out, this effect can lead to both an increase and a decrease in the growth rate of the particles depending on the vapor saturation ratio. In [12] the problem of the influence of radiative heating of aerosol particles with adsorbed molecules of a buffer gas on the resultant flux of vapor molecules into the particle (and correspondingly on its growth rate) was investigated.

We will consider the problem on the mutual influence exerted on the critical particle size by an electric charge and adsorption of the molecules of a foreign gas that decreases the surface-tension coefficient. We will use the expression given in [13] for the dependence of the surface-tension coefficient on the pressure of an adsorbable gas P_a:

$$\sigma(P_a) = \sigma(0) - n_0 kT \ln\left(\frac{P_a + P_L}{P_L}\right). \tag{15}$$

With allowance for Eqs. (2)–(5) and (15), we obtain the following expression for the critical particle diameter at $Q_3 = 0$:

$$\ln S = \frac{V_m}{kT}\left[\frac{4\sigma(0)}{d_{cr}} - \frac{2q^2}{\pi d_{cr}^4}\left(\frac{1}{\varepsilon_g} - \frac{1}{\varepsilon_c}\right)\right] + \frac{4V_m n_0}{d_{cr}} \ln\frac{P_L}{P_a + P_L} - \ln\frac{\alpha_c}{\alpha_e}. \tag{16}$$

As shown by calculations, the presence of a foreign adsorbable gas in a system leads to a change in the critical particle diameter. Here, with increase in the pressure of an admixture gas a smaller value of S is needed that results in the realization of a definite value of the critical particle size. The difference in the condensation coefficient from the evaporation coefficient (for $\alpha_c < \alpha_e$) leads to a certain compensation of the effect, which is connected with the presence of a foreign adsorbable gas in the system. The change in the critical particle size caused both by the effect due to the dependence of the condensation coefficient on the particle size and by the action of electromagnetic fields can substantially influence the rate of homogeneous nucleation because the latter is very sensitive to a change in the critical cluster size. The nucleation rate in a general form can be written as follows [6]:

$$J = C \exp\left(-\frac{G}{kT}\right), \tag{17}$$

where the free energy of the critical cluster generation G depends on the cluster size and the preexponential term C includes the condensation coefficient.

At high enough pressures of adsorbable gas, polymolecular films of an adsorbate substance can be formed on nanosized particles. In this case, a critical film thickness can be realized at which the resultant influx of vapor molecules into the film is equal to zero. The study of the laws governing the deposition of polymolecular films on aerosol particles is very topical for atmospheric processes related to generation of water films on particles and for the processes of modern technology, for example, in obtaining nanoparticles coated with a film of another substance (in particular, such coatings are used in heterogeneous catalysis to raise the catalytic ability of particles). The thickness of the film formed depends on the size and substance of the particles, the saturation ratio of the substance deposited, the presence, in the gas phase, of foreign gas molecules that can be adsorbed on the film surface and change the surface-tension coefficient, as well as on the influence of external fields (for example, resonance radiation). In [14], some problems connected with the determination of the critical (equilibrium) thickness of a liquid polymolecular wetting film on an aerosol particle in vapor condensation are considered. Expressions for the critical film thickness on a small spherical particle for nonpolar and polar molecules of the film substance in the presence of molecules of a buffer (admixture) gas that are adsorbed on the film surface were derived. As follows from the results obtained, the value of the critical film thickness increases with the particle diameter and the pressure of the foreign gas that is adsorbed on the film surface and that decreases the surface-tension coefficient.

Critical Size of Aerosol Particles in Chemical Deposition. Above, some problems related to the critical size of aerosol particles in physical deposition (vapor condensation) were considered. In many phenomena occurring both in the atmosphere and in diverse technological processes, chemical deposition of a substance takes place. In a number of works devoted to such a type of deposition, the reverse flow of molecules from the condensed phase formed is not taken into account; thus, it is implicitly supposed that the equilibrium condition (equality to zero of the resultant mass flux into the condensed phase) does not hold in the system considered. The question arises as to the possibility of realization of the critical size of aerosol particles in chemical substance deposition, and it is this question that will be considered below.

It is known that deposition of molecules on the surface of aerosol particles can occur in a heterogeneous chemical reaction of gas molecules on the surface of a particle with the formation of molecules (atoms), from which a condensed phase is formed, and fragments of initial molecules that desorb back into the gas phase. An initial gas molecule can also pass into a chemisorbed state as a whole. The processes of chemical deposition play a very important role both for the physicochemistry of the Earth's atmosphere and for obtaining nanoparticles. We note that in the latter case first the chemical reaction of decomposition of an initial substance (precursor) in a gas phase is realized, with the resulting appearance of the component whose molecules, in turn, are able to participate in the process of nucleation and growth of the clusters originated.

Further we will consider chemical deposition on already available aerosol particles. To describe the processes mentioned, we will use the following simple scheme of chemical deposition. We consider that the molecules of gas-reactant (precursor) AB collide with the particle surface, with the possible occurrence of a chemical reaction that leads to the transition of one of its products into a bound state (c) (a condensed phase or a state of chemical adsorption) and to generation of gas molecules (g) that desorb from the surface:

$$\mathrm{AB\ (g)} \rightarrow \mathrm{A\ (c)} + \mathrm{B\ (g)}\ . \tag{18}$$

We will analyze the case where a reaction of type (18) occurs with adsorption of reactant molecules on the particle surface. For simplicity, the sticking and evaporation coefficients of molecules (atoms) are supposed to be equal to unity. Further the surface coverage by the adsorbed molecules is assumed to be well below unity, and the number density of molecules (atoms) of component A at an infinite distance from the particle is considered zero. Here, within the framework of the assumptions accepted in [15], we can write the following expression for the density of the resultant flux of molecules (atoms) of component A into the particle:

$$j_{\mathrm{A}} = \frac{\left(1+\dfrac{k_{\mathrm{d}}}{k_{\mathrm{r}}}\right)^{-1} \dfrac{n_{\mathrm{AB}\infty} v_{\mathrm{AB}}}{4}}{1+\left(1+\dfrac{k_{\mathrm{d}}}{k_{\mathrm{r}}}\right)^{-1} \dfrac{R v_{\mathrm{AB}}}{4D_{\mathrm{AB}}}} - \frac{\dfrac{n_{\mathrm{Ae}} v_{\mathrm{A}}}{4} \exp\left(\dfrac{2\sigma V_{\mathrm{A}}}{RkT}\right)}{1+\dfrac{R v_{\mathrm{A}}}{4D_{\mathrm{A}}}}, \tag{19}$$

where k_{r} and k_{d} are respectively the rate constants of the heterogeneous chemical reaction and desorption of gas-reactant molecules from the particle surface:

$$k_{\mathrm{d}} = k_{\mathrm{d0}} \exp\left(-\frac{Q_{\mathrm{d}}}{kT}\right), \quad k_{\mathrm{r}} = k_{\mathrm{r0}} \exp\left(-\frac{Q_{\mathrm{r}}}{kT}\right). \tag{20}$$

It is seen from (19) that in chemical deposition the critical particle size (which is determined by the equality $j_{\mathrm{A}} = 0$) generally depends on the diffusion coefficients of molecules of reactant AB and molecules (atoms) A evaporating from the particle. In a free-molecule regime of a gas flow, where the mean free path of molecules (atoms) greatly exceeds the particle radius (i.e., with $Rv_{\mathrm{AB}}/(4D_{\mathrm{AB}}) \ll 1$ and $Rv_{\mathrm{A}}/(4D_{\mathrm{AB}}) \ll 1$), the expression for the critical particle diameter can be written as

$$d_{\mathrm{cr}} = \frac{4\sigma V_{\mathrm{A}}}{kT \ln \dfrac{n_{\mathrm{AB}\infty} v_{\mathrm{AB}}}{\left(1+\dfrac{k_{\mathrm{d}}}{k_{\mathrm{r}}}\right) n_{\mathrm{Ae}} v_{\mathrm{A}}}}\,. \tag{21}$$

It follows from (21) that the critical diameter of the particles that are formed in chemical deposition decreases with increase in the number density of reactant molecules and with decrease in that for the molecules (atoms) of the saturated vapor of the particle substance, as well as with increase in the ratio $k_{\mathrm{r}}/k_{\mathrm{d}}$.

Conclusions. The problem of the critical size of aerosol particles in the case of physical and chemical deposition has been considered. It is shown that the critical particle diameter can experience the influence of such factors as the dependence of the condensation coefficient on the particle size, the presence of a foreign adsorbable gas in the system, and the action of an electromagnetic field. The problem of the critical size of aerosol particles in chemical deposition has been discussed.

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NOTATION

D, D_{AB}, and D_A, diffusion coefficients of molecules of vapor, reactant, and component A in a buffer gas, respectively; d_{cr}, critical particle diameter; G, free energy of cluster formation; h, Planck constant; I_ν, radiation flux density; k, Boltzmann constant; k_{d0} and k_{r0}, preexponential factors for the rate constants of desorption and heterogeneous chemical reaction; m_a, mass of a molecule of a foreign adsorbable gas; n_0, number of adsorption centers per unit surface; n, number density of vapor molecules; $n(R)$, number density of vapor molecules at the particle surface; n_e, number density of molecules of saturated vapor over a flat surface; n_{AB}, number density of reactant molecules; n_{Ae}, number density of molecules of saturated vapor of component A over a flat surface; Q_d and Q_r, desorption heat of reactant molecules and activation energy of heterogeneous chemical reaction, respectively; q, elementary electric charge; P_a, pressure of a buffer (admixture) gas; R, particle radius; S, saturation ratio; T, temperature (for simplicity the isothermal case is considered); v_{AB}, mean velocity of reactant molecules; v_A, mean velocity of molecules (atoms) of component A; v, mean velocity of vapor molecules; V_m, volume per molecule in a particle; V_A, volume per molecule (atom) of component A in a particle; α_c and α_e, condensation and evaporation coefficients for a clean surface, respectively; α_A, condensation coefficient of component A which is supposed to be equal to evaporation coefficient; β, coefficient of sticking of a molecule of adsorbable gas to the surface region which is not occupied by adsorbed molecules; ε_g and ε_c, dielectric constants of gas and condensed phase; θ, surface coverage by molecules of foreign adsorbable gas; ν, radiation frequency; σ, surface-tension coefficient; $\sigma(0)$, surface-tension coefficient of a clean liquid; σ_ν, resonance absorption section; τ, adsorption time of molecules of a foreign gas; τ_ν, relaxation time of excited gas molecules. Subscripts: a, adsorption; c, condensation; cr, critical; d, desorption; e, evaporation; g, gas; m, molecule; r, reaction; ∞, infinite distance from a particle.

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