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# INFLUENCE OF SURFACE EFFECTS IN VAPOR CONDENSATION ON A NANODIMENSIONAL AEROSOL PARTICLE HAVING AN ELECTRIC CHARGE

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*The influence of surface phenomena and size effects on vapor condensation on a charged aerosol particle is considered.*

The processes of nucleation and growth of particles in deposition from a gas phase are of high current interest both in investigation of atmospheric phenomena (for example, generation and growth of water drops as well as smog particles) and in different fields of new technology connected with obtaining and using nanoparticles. Furthermore, questions arise as to the influence of different effects on the formation and growth of particles of a condensed phase. It is known that in a number of cases, along with neutral particles, a gas phase can contain particles which possess an electric charge. For example, in the Earth's atmosphere there are a fairly large quantity of ions which appear not only under the action of cosmic radiation but also as a result of different technological processes. On the other hand, the presence of charged particles in a gas phase facilitates initiation of the process of homogeneous nucleation [1, 2]. Here, the influence of a charge can manifest itself through different mechanisms. For example, it is shown in [3, 4] that under the action of a charge the distribution of the concentration of polar vapor molecules near the particle is changed, which leads to a change in the critical size of particles and in the flux of vapor molecules into the particle. In [5], a thermodynamic approach is used to investigate the action of the particle charge on the surface tension, and it is shown that this action influences the phase transition on the particle surface and homogeneous nucleation. Below, we consider the influence of the factors which change the condensation coefficient of polar vapor molecules on phase transformations on the surface of charged aerosol particles (drops) and on their critical (equilibrium) size. In so doing, we ignore, for simplicity, the above-mentioned effects connected with the change in the vapor concentration near the particle as well as with the change in the surface-tension coefficient under the action of an electric field.

We shall consider a small aerosol particle with an electric charge $q$ at its center. Hereinafter, we shall use the so-called drop model in which a particle of arbitrary size is considered as a macroscopic drop. In so doing, we suppose that the concept of surface tension can also be applied to fairly small particles (clusters). It is clear that the use of the surface-tension coefficient for small clusters is the subject of discussion. However, as is known, the drop model is applied in classical nucleation theory to arbitrary-size particles. In [6], the surface-tension coefficient for particles of fairly small diameters is also considered.

In studying problems of phase transformations on charged particles, of high importance is the value of the critical (equilibrium) size of particles. For example, the rate of homogeneous nucleation depends heavily on this value. The saturation ratio corresponding to the critical diameter of a particle (with an electric charge at the center) is usually calculated on the basis of the equation [2]

$$\ln S = \frac{M}{\rho RT}\left[\frac{4\sigma}{d_{\text{cr}}} - \frac{2q^2}{\pi d_{\text{cr}}^4}\left(\frac{1}{\varepsilon_{\text{g}}} - \frac{1}{\varepsilon_{\text{liq}}}\right)\right] \quad (1)$$

(here and hereafter, the CGS electromagnetic system of units accepted in [2] is used).

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We note that expression (1) does not contain the condensation coefficient. Since the above equation, which relates the saturation ratio to the critical particle diameter, contains the squared quantity $q$, the value of the critical diameter should be independent of the charge sign (at equal values of other parameters). It follows from this that there is no dependence of the nucleation rate on the sign of $q$. However, experiments have revealed such a dependence (a review of works devoted to the dependence of the nucleation rate on the charge sign can be found, for example, in [7, 8]). Thus, for water condensation on a negatively charged particle smaller vapor supersaturation is required than in the case of a positively charged one [2]. This dependence can appear with an asymmetry in the interaction of polar vapor molecules with the particles that possess charges of different signs, which, in particular, can be explained by the difference in the condensation coefficients for polar molecules incident on the aerosol particles having different charges. This is connected with the following factor. Owing to the fairly small time of rotational relaxation [9], the polar molecules (dipoles) impinging on a charged particle will be oriented in a definite way relative to the particle surface, depending on the charge sign. At the same time, incorporation of the molecules incident on the particle into the condensed phase may depend on how (by which dipole end) the given molecule is oriented relative to the surface of the condensed phase. For example, water molecules are more easily condensed on the aerosol particles having a negative charge (when the electric field orients dipoles by their positive charge directed to the particle surface). We note that the calculations carried out in [10] for the adsorption of a water molecule on the end section of a carbon nanotube without a charge showed that the situation appeared to be more stable when this molecule faces the nanotube surface by its positive dipole charge. The condensation (sticking) coefficient can also depend on excitation of the internal degrees of freedom of molecules (for example, in the field of resonance laser radiation), which affects the value of $d_{\mathrm{cr}}$ [11]. Moreover, generally the condensation coefficient depends also on the particle size [12–14]. In [13], the process of molecule condensation on a small aerosol particle is considered from the point of view of coagulation of two particles, one of which is the molecule. Here, too, a conclusion is drawn regarding the dependence of the condensation coefficient on the size of an aerosol particle. All the above suggests that in the general case the critical particle size (and, correspondingly, the rate of homogeneous nucleation) will depend on the character of interaction of a gas molecule with the particle surface, which is characterized by the condensation coefficient.

We will determine the resulting flux of vapor molecules into a particle having an electric charge. The influence of the effects due to the surface curvature and to the presence of the particle charge on the evaporation intensity can be connected with the change in the energy of evaporation of molecules from the particle [15]. Further, for simplicity, we shall take into account only the electrostatic energy of interaction of polar molecules with the charged particle (this corresponds to the approximation in which expression (1) was obtained), and the interaction energy related to the polarization of molecules in an electric field will be ignored (it can be allowed for in the same way as in [15]).

Taking into account the blocking influence of the foreign (buffer) gas molecules adsorbed on the particle surface, subject to the foregoing and [16, 17], we obtain the following expression for the density of the resulting flux of molecules into the particle in a free-molecular regime of gas flow under the assumption of the Maxwellian velocity distribution function of molecules:

$$I = (1-\theta)\,\alpha_{\mathrm{c}}\frac{P}{(2\pi mkT_{\mathrm{g}})^{1/2}} - (1-\theta)\,\alpha_{\mathrm{e}}\frac{P_{\mathrm{e}}(T_{\mathrm{p}})}{(2\pi mkT_{\mathrm{p}})^{1/2}}\exp\left\{\frac{M}{\rho RT_{\mathrm{p}}}\left[\frac{4\sigma}{d} - \frac{2q^2}{\pi d^4}\left(\frac{1}{\varepsilon_{\mathrm{g}}} - \frac{1}{\varepsilon_{\mathrm{liq}}}\right)\right]\right\}. \tag{2}$$

The difference in the values of $T_{\mathrm{g}}$ and $T_{\mathrm{p}}$ (the particle heating is assumed to be uniform) can be due to both the generation of the latent heat of phase transition in condensation of vapor molecules and to the particle heating by incident radiation.

It should be noted that if a foreign gas in which condensation occurs consists of polar molecules, the electric charge present in an aerosol particle can enhance adsorption of these molecules on the particle (cluster) surface. These molecules will obstruct the phase-transition surface to the molecules condensed. Thus, using the Langmuir model of monomolecular adsorption, for $1-\theta$ we have

$$1-\theta = \frac{1}{1+P_{\mathrm{a}}/P_0}, \tag{3}$$

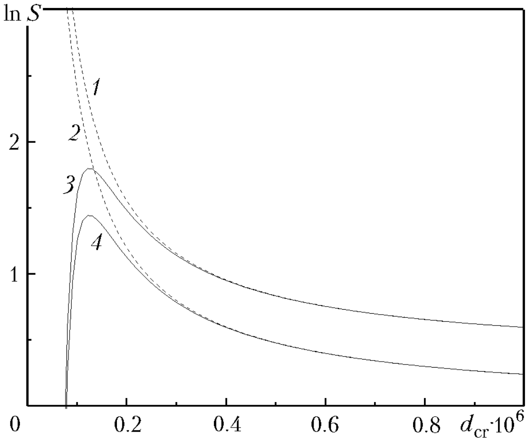

Fig. 1. Dependence of ln $S$ on the critical diameter for a neutral water drop (dashed curves) and a drop with a unit charge (solid curves) at $T$ = 273 K: 1, 3) $\alpha_c/\alpha_e$ = 0.7; 2, 4) $\alpha_c/\alpha_e$ = 1. $d_{\rm cr}$, cm.

where $P_a$ is the partial pressure of the foreign polar gas, and, taking into account the action of an electric field on polar molecules, the quantity $P_0$ can be presented as

$$P_0 = \frac{n_0 (2\pi m_a k T_g)^{1/2} \exp\left\{-\frac{Q_a}{RT_p} - \frac{M_a}{\rho_a R T_p}\left[\frac{2q^2}{\pi d^4}\left(\frac{1}{\varepsilon_g} - \frac{1}{\varepsilon_{\rm liq}}\right)\right]\right\}}{\beta\tau_0}. \tag{4}$$

It is seen from relations (2)–(4) that the presence of a charge leads to a decrease in the value of $P_0$ and, correspondingly, in the value of $1 - \theta$. Thus, a situation is possible, in principle, where the charge of an aerosol particle will assist adsorption of molecules of a foreign gas, which will decrease the resulting flux of vapor molecules into the particle due to the blocking effect of the molecules adsorbed. At a sufficiently high pressure of a foreign gas, formation of polymolecular film consisting of the molecules of this gas can begin on the particle surface. Certain aspects of deposition of polymolecular films on aerosol particles (in particular, critical film thickness) are considered in [18].

It should be noted that the presence of adsorbed molecules of a foreign gas on the particle surface can change the surface-tension coefficient as well. The joint influence of the blocking action of the molecules adsorbed on the particle surface and of the dependence of the surface-tension coefficient on the surface coverage by these molecules for a neutral particle is considered in [17].

For the value of the critical particle size determined by the condition $I$ = 0, from (2) we have

$$\ln\left[\frac{\alpha_c}{\alpha_e}\frac{P}{P_e(T_p)}\sqrt{\frac{T_p}{T_g}}\right] = \frac{M}{\rho R T_p}\left[\frac{4\sigma}{d_{\rm cr}} - \frac{2q^2}{\pi d_{\rm cr}^4}\left(\frac{1}{\varepsilon_g} - \frac{1}{\varepsilon_{\rm liq}}\right)\right]. \tag{5}$$

It follows from (5) that generally the critical particle diameter depends on both the value of the condensation coefficient and the particle and gas temperatures. For simplicity, further we shall consider an isothermal case ($T_g = T_p = T$). In so doing, from (5) we have

$$\ln S = \frac{M}{\rho R T}\left[\frac{4\sigma}{d_{\rm cr}} - \frac{2q^2}{\pi d_{\rm cr}^4}\left(\frac{1}{\varepsilon_g} - \frac{1}{\varepsilon_{\rm liq}}\right)\right] - \ln\left(\frac{\alpha_c}{\alpha_e}\right). \tag{6}$$

At $\alpha_c = \alpha_e$, expression (6) goes over into (2).

We consider the influence of the saturation ratio on the critical particle (drop) diameter at different values of $\alpha_c/\alpha_e$. In so doing, we reason, for simplicity, that the value of the surface-tension coefficient is independent of the particle size and is equal to its value for a volumetric phase (the dependence of this coefficient on the particle size will be analyzed below). Figure 1 shows the dependence of ln $S$ on the critical diameter of a drop with a unit elementary charge for different values of the parameter $\eta = \alpha_c/\alpha_e$. As follows from the figure, within increase in $\eta$, the critical particle diameter decreases at the same values of the saturation ratio. In turn, this leads to an increase in the rate of homogeneous nucleation, because the latter increases with a decrease in the critical size of the clusters.

It should be noted that the condensation and surface-tension coefficients for sufficiently small particles depend, generally, on their diameter $d$, and the influence of the particle size on the evaporation probability is taken into account by the exponent in expression (2) (the evaporation coefficient $\alpha_e$ can be considered independent of $d$).

According to [12] and [19], the dependences $\alpha_c(d)$ and $\sigma(d)$ can be approximated by the following expressions:

$$\alpha_c = \alpha_{c,f} \exp\left(-\frac{6\sigma M}{d\rho RT}\right), \tag{7}$$

$$\sigma(d) = \frac{\sigma_f}{1 + \dfrac{4\delta}{d}}, \tag{8}$$

where $\alpha_{c,f}$ and $\sigma_f$ are the condensation and surface-tension coefficients for a flat surface; $\delta$ is the so-called Tolman length for which the range of change 0.096–0.35 nm is given in [19] (in the present work, the value $\delta = 0.2$ nm is used in calculations). We note that derivation of the Tolman formula (8) was performed on the assumption that the value of $2\delta/d$ is small as compared to unity. However, the comparison carried out in [6, 20] between the results of calculations of the surface-tension coefficient for small particles (down to the values of the parameter $2\delta/d$ of the order of unity) on the basis of different models, as well as by computer simulation, and the results following from the Tolman formula enables one to conclude that the formula can be used for drops of very small diameter.

In the case of using expressions (7) and (8) for $\alpha_c$ and $\sigma$, on the assumption that $\alpha_{c,f} = \alpha_e$, from (6) we obtain

$$\ln S = \frac{M}{\rho R T_p}\left[\frac{10\sigma_f}{d_{cr} + 4\delta} - \frac{2q^2}{\pi d_{cr}^4}\left(\frac{1}{\varepsilon_g} - \frac{1}{\varepsilon_{liq}}\right)\right]. \tag{9}$$

We shall find the resulting flux of vapor molecules into a particle at arbitrary Knudsen numbers. In the general case of a drift of polar vapor molecules in an external field, the density of the flux of molecules onto the particle surface will consist of diffusive and convective components. The latter is connected with the appearance of a force which acts on polar molecules in the field of a charged particle [15]. However, next we consider, as in [15], that the distribution of the concentration of vapor molecules in the vicinity of the particle in the quasi-stationary approximation can be described by the expression corresponding to the steady-state solution of the equation of the diffusion of vapor molecules in a buffer gas without regard for the convective component of the flux of molecules. Taking into account the foregoing together with (5), (6), and work [21], in the isothermal approximation for arbitrary Knudsen numbers we obtain the following expression for the density of the resulting flux of molecules into a drop:

$$I = \frac{\dfrac{(1-\theta)P_\infty}{(2\pi mkT)^{1/2}}\left[\alpha_{c,f}\exp\left\{-\dfrac{M}{\rho RT}\dfrac{6\sigma_f}{d+4\delta}\right\} - \dfrac{P_e}{P_\infty}\alpha_e \exp\left\{\dfrac{M}{\rho RT}\left[\dfrac{4\sigma_f}{d+4\delta} - \dfrac{2q^2}{\pi d^4}\left(\dfrac{1}{\varepsilon_g} - \dfrac{1}{\varepsilon_{liq}}\right)\right]\right\}\right]}{1 + (1-\theta)\,\alpha_{c,f}\exp\left\{-\dfrac{M}{\rho RT}\dfrac{6\sigma_f}{d+4\delta}\right\}\dfrac{\nu d}{8D}}, \tag{10}$$

where $\dfrac{\nu d}{8D} = \dfrac{3}{4\mathrm{Kn}}$; $\mathrm{Kn} = 2\lambda/d$.

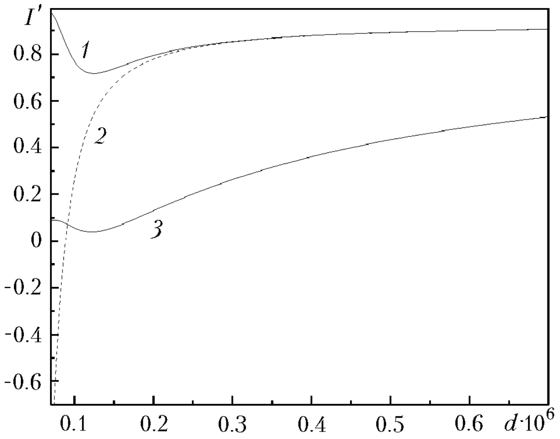

Fig. 2. Dependence of the dimensionless density of the resulting flux of vapor molecules into a neutral water drop (dashed curve) and a drop with a unit charge (solid curves) on the drop diameter at $T = 273$ K, $\delta = 0.2$ nm, $P_\infty/P_e = 15$: 1, 2) $\alpha_c = \alpha_{c,f}$, $\sigma = \sigma_f$; 3) $\alpha_c = \alpha_c(d)$, $\sigma = \sigma(d)$. $d$, cm.

We consider the influence of the size of particles on the resulting flux of vapor molecules into a particle. Further we suppose, for simplicity, that $\theta = 0$ and $\alpha_{c,f} = \alpha_e = \alpha$. Figure 2 shows the dimensionless density of the flux of vapor molecules into a water drop $I' = I(2\pi mkT)^{1/2}/(\alpha P_\infty)$ in the region with Kn >> 1 with allowance made for the influence of the charge as well as of the surface effects connected with the dependence of the values of $\alpha_c$ and $\sigma$ on the drop size. For comparison, the figure also gives the flux density of vapor molecules for a neutral drop with constant condensation and surface-tension coefficients. It is seen from Fig. 2 that the presence of a charge leads to the appearance of a minimum in the size dependence of the density of the resulting flux of molecules into the drop. This is associated with the competition of the effects acting in the opposite directions on increase in the drop size. Thus, with an increase in the drop size the influence of the charge of a drop, which decreases the reevaporation of molecules from it, results in a fall in the density of the resulting flux of molecules. At the same time, as the size of the particle increases, the condensation coefficient increases and the Kelvin effect manifests itself more weakly, which increases the density of the resulting vapor flux into the particle. We note that, as follows from Eq. (10), beginning from a definite drop size, the influence of the surface processes on the density of the resulting flux of vapor molecules into a drop decreases, and the main part is now played by the intermolecular-collisions-related resistance to the delivery of molecules to the drop (in the case of its growth) or to their outflow (during its evaporation). Here, the density of the resulting flux of molecules into the drop will decrease with an increase in its diameter. It should be noted that the case where a charge is at the particle center was considered above.

In the case of an ion being adsorbed on the surface of a small particle, effects that are connected that the nonuniformity of the intensity of phase transitions on its surface may occur. As is seen from Fig. 2, the influence of a charge on the resulting flux of molecules manifests itself for fairly small diameters of particles (i.e., the distances from the charge to the surface of phase transition). In the presence of a charge on the particle surface, its influence on phase transition will show itself to a greater extent on that side of the particle where the charge is present. Here, a new component of the force acting on the particle can appear due to the fact that the density of the resulting flux of vapor molecules on the given particle side will be higher than on the opposite side. The appearance of the indicated force will also affect the particle motion in a gas phase. When a particle with a charge on the surface is situated in an external electric field, orientation of the drop–charge system relative to the field can take place (the charge on the particle will be oriented towards the oppositely charged source of the external field). The situation is analogous to the orientation of dipoles in an external electric field. If there are several ions on the particle surface, then under the effect of the external field they can be concentrated on one of the particle sides as a result of their drift over the surface due to the field. In condensation growth of a particle with an asymmetrical charge on the surface, in an external electrical field this particle will experience the action of both the electrophoretic force connected with the interaction between the external field and the charge and the above-mentioned phoretic force caused by the nonuniformity of phase transitions on the particle surface due to the uniform charge distribution. Here, the increase in the resulting flux of vapor molecules on the particle side having a higher charge density is generally caused by both the increase in the effective energy of evaporation of the polar molecule from the drop and the higher value of the density of the flux of polar molecules incident on this side of the drop. The second factor of those mentioned is related to the action of the electric charge on the gas phase [3].

Thus, in this work the process of condensation of polar vapor molecules on a small charged particle (drop) has been considered with allowance made for the size effects connected with the dependence of the condensation and surface-tension coefficients on the drop size. The difference between the condensation coefficients of polar water molecules in the case of their different orientation relative to the drop surface may be one reason for the change in the value of the saturation ratio on change in the sign of the drop charge. In turn, this leads to different values of the rate of homogeneous nucleation for positively and negatively charged clusters. The possibility of the appearance of an additional force acting on a charged aerosol particle which is placed in an electric field and which grows there during vapor condensation when a charge is found on its surface is discussed. The appearance of this force is attributable to the nonuniform distribution of the density of the resulting flux of vapor molecules over the particle surface due to the nonuniformity of the charge distribution over the surface.

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## NOTATION

$D$, coefficient of vapor diffusion in a buffer gas; $d$, particle diameter; $d_{\mathrm{cr}}$, critical diameter of a particle; $I$, density of the resulting flux of vapor molecules into a particle; $k$, Boltzmann constant; Kn, Knudsen number; $M$, molecular weight; $m_{\mathrm{a}}$, mass of a molecule of adsorbing gas; $n_0$, number of adsorption centers on a unit surface; $P$, partial vapor pressure in a gas phase; $P_{\mathrm{e}}$, saturation vapor pressure over a flat surface; $Q_{\mathrm{a}}$, adsorption energy for a neutral particle; $q$, charge; $R$, universal gas constant; $S$, saturation ratio; $T$, temperature; $v$, thermal velocity of vapor molecules; $\alpha_{\mathrm{c}}$, coefficient of molecule condensation on the part of the particle surface not covered by adsorbed molecules of a foreign gas; $\alpha_{\mathrm{e}}$, evaporation coefficient for the clean part of the particle surface; $\beta$, coefficient of sticking of a molecule of adsorbing gas to the surface part not occupied by adsorbed molecules; $\varepsilon_{\mathrm{g}}$ and $\varepsilon_{\mathrm{liq}}$, dielectric constants of gas and liquid; $\theta$, surface coverage by adsorbed molecules of a foreign (buffer or admixture) gas; $\lambda$, free path length of vapor molecules in a buffer gas; $\sigma$, surface-tension coefficient; $\rho$, density; $\tau_0$, quantity characterizing the oscillation period of an adsorbed molecule in the direction normal to the surface. Subscripts; a, adsorption; c, condensation; cr, critical; e, evaporation; f, flat; g, gas; p, particle; $\infty$, infinite distance away from a particle; liq, liquid.

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