Measurement of the drag force for charged microparticles by the electrodynamic levitation method

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The results of drag force measurements by the EDL method in air for lycopodium particles of 28-µm diameter with the Knudsen numbers ranging from 0.1 to 3.0 are presented. The EDL design with parallel plate electrodes used in the experiments is described, as well as the measurement method based on recording the microparticle leaving the stability curve. The results obtained are compared with the molecular-kinetic theory for the effect under study, and their good agreement in the used range of Knudsen numbers is found at the tangential momentum accommodation coefficient from 0.8 to 1.0. The problems concerning the Brownian motion of particles at low gas pressures and the influence of a surface particle charge on the drag force are considered.

Introduction

In Ref. 1 we described a laboratory facility for studying microphysical characteristics of aerosol particles – the electrodynamic levitation (EDL) with plane-parallel electrodes placed in a vacuum chamber. It is one of the possible versions of such devices² that has some advantages over EDL with other electrode configurations.¹ In contrast to the most of other EDL modifications, this device allows the measurements to be carried out at low gas pressure and different intensity of radiation incident on microparticles. This technique is promising, in particular, for studying the processes of interaction of the electromagnetic radiation with particles from various substances in a wide size range $(1-100 \,\mu\text{m})$ at different pressure of the ambient gas (0.01-760 Torr). Apparently, such experimental capabilities allow modeling the behavior of real atmospheric aerosols at different altitudes (up to 80-90 km) in the field of solar radiation and the outgoing Earth's radiation.

Such modeling of dynamic processes with stratospheric aerosol calls for confirmation of the efficiency and accuracy of the proposed measurement methods (in particular, the method of achieving the particle's stability threshold) and validity of the results obtained with the use of a particular experimental setup. The obvious "geometric" test of such a kind is the determination of size (aerodynamic diameter) of the particles under study and comparison of the results with the data of optical microscopy. As a dynamic, more sensitive test, we can propose determination of the drag force of spherical microparticles with the known properties and size in a wide range of the ambient gas pressure. As will be shown below, the results of this kind are beyond the frames of standard test measurements and are characterized by sufficient scientific novelty and originality.

Determination of the drag force of a spherical particle in a gas flow depending on the Reynolds and

Knudsen numbers is a basic problem of the kinetic theory of rarefied gases and mechanics of aerosols. Theoretically, this problem has been studied thoroughly by now, which allows us to use the reliable and high-accuracy theoretical data obtained, in particular, based on ideas of molecular-kinetic theory^{4,5} for a comparison with the experimental data.

Experimental investigations of the drag force in a wide range of the Knudsen number go back to the well-known results by Millikan (see Ref. 2 and references therein) for clocks oil droplets in air and other gases. Current investigations carried out by the EDL technique are mostly devoted to studying the dependence of the particles' drag force on the Reynolds number (at the fixed (small) value of the Knudsen number) and to determining the dynamic shape factor at motion of nonspherical particles. To be noted here are the results from Ref. 6, in which the quadrupole EDL was connected with a miniature aerodynamic nozzle for generation of a laminar gas flow over microparticles. In the experiments, the Reynolds number ranged within Re = 0-5, and the measurement findings agreed very well with the theoretical predictions. The dependence of the drag force on the Knudsen number was not investigated, since the measurements were conducted at atmospheric pressure (the measuring cell was not evacuated), and all the used particles had almost equal size.

In Refs. 7 and 8, a new version of the informative method for determination of aerodynamic particle diameters, drag force, and other microphysical parameters was proposed for the EDL configuration with double ring electrodes, which was used for the first time in Ref. 9 for other purposes. Note that independent of Refs. 7 and 8, we have developed similar technique and used it (see Ref. 1). It is based on electric measurements of the characteristics of direct and alternating electromagnetic fields at the stability threshold of motion of a charged particle (the amplitude of particle oscillations increases sharply in this case, and this can be fixed with high accuracy). This method was called the spring-point method.⁸ Since measurements in Refs. 7 and 8 were conducted only at the atmospheric pressure as well, the dependence of the particles' drag force on the Knudsen number could not be investigated. In this paper, we present, for the first time, the results of such measurements and their tentative analysis.

Technique for determination of the drag force

The equation of motion of a charged microparticle along the axis z in the EDL volume (Fig. 1) has the following form:

$$m\frac{d^{2}z}{dt^{2}} + F_{U}(Kn)\frac{dz}{dt} - qE_{ac} = q[E_{dc} - E_{dc}(0)], \quad (1)$$

where *m* is the particle mass; η is the dynamic viscosity of the ambient gas; *q* is the particle charge; *t* is time; $E_{\rm ac}$ and $E_{\rm dc}$ are the strength of ac and dc electric fields in the EDL volume; $E_{\rm dc}(0)$ is the strength of the dc electric field holding the particle at the EDL center with the force of gravity equal to the Coulomb force $qE_{\rm dc}(0) = mg$ (*g* is the acceleration due to gravity); $F_U({\rm Kn})$ is the isothermal drag force of gas rarefaction (Knudsen number ${\rm Kn} = l/R_{\rm p}$, where *l* is the mean free path of gas molecules determined from the gas viscosity; $R_{\rm p}$ is the particle radius). For a large particle at small Re, it corresponds to the Stokes law, and for other gas-kinetic conditions it is calculated based on the molecular-kinetic theory.^{4,5}



Fig. 1. EDL with plane-parallel ring and disc electrodes $(d = 1 \text{ mm}, R_0 = 6.7 \text{ mm}, \text{ copper wire; } 2h = 4 \text{ mm}; 2H = 30 \text{ mm}; D_0 = 60 \text{ mm}, \text{aluminum discs}).$

Taking into account the form of solution of Laplace equation for the potential generated by the electric fields in the EDL volume of a given configuration (see, for example, Ref. 10), we can reduce Eq. (1) to the Mathieu equation describing nonlinear oscillation processes in the electrical and mechanical systems. This formalism is described in detail in Refs. 1 and 8. Depending on the values of the viscous drag parameter $A = F_U(\text{Kn})/m\omega$ (ω is the circular frequency of the ac voltage) and the electric field parameter $B = C_{ac}gV_{ac}/C_0\omega^2V_{dc}(0)$ (C_{ac} and C_0 are geometric constants determined by the size and shape of the electrodes), the solution of the Mathieu

equation describes both stable and unstable modes of the particle motion in the EDL volume. The curve separating these regions in the coordinates A, B is called the stability threshold and it is very important characteristic of the EDL of any type. For the first time, the approximate equation of this curve for the electrode configuration used was obtained in Ref. 11.

In Ref. 1, the approximate analytical solution of this problem was obtained by the Bubnov–Galerkin method and studied numerically by the finitedifference method. Figure 2 depicts the data of theoretical analysis of the stability threshold curve corresponding to approximate solution¹¹ and numerical analysis.¹ Measurements of the electric parameters (voltage and frequency) of particle leaving from this curve can be used directly to get the information about the drag force.



Fig. 2. Stability threshold curve for particle motion in the EDL volume: numerical solution by the finite-difference method¹ (t) and approximate analytical solution¹¹ (2).

The particular measurement technique was the following. A small number of particles having gained a charge in the ionizer fell in the working chamber (see Fig. 1). As this took place, only ac voltage was applied $V_{\rm ac}$ to the ring electrodes. A part of falling particles was trapped by the ac electric field in different places between the ring electrodes. Then, decreasing $V_{\rm ac}$, we got that all particles but one deposited on the electrodes or the chamber walls. The minimum possible value of $V_{\rm ac}$, at which the gas breakdown in the chamber at low pressure still did not occur and the particle did not leave the interelectrode zone, was about 300 V.

Adjusting and applying the dc voltage $V_{dc}(0)$ to the disc electrodes, we can place the particle just at the EDL center, where it can be in the steady state (lying on the stability threshold curve) for a long time at a certain value of V_{ac} . Changing the values of V_{ac} and/or ω , we can achieve the unsteady state, at which the particle oscillates intensely, that is, moves in the immovable gas in the chamber. The electrical parameters corresponding to particle leaving from the stability threshold curve can be measured with very high accuracy. Gradually evacuating the chamber, we determined the sets of $V_{\rm ac}$ and ω values at a fixed gas pressure that corresponded to the loss in stability of the particle motion at different Knudsen numbers. The gas pressure was decreased until the increasing intensity of the Brownian motion of the particle that yet allowed such measurements to be conducted. Then, using the results of solution of the equation for the stability threshold curve, we determined the drag force of particles of a particular substance and size at different gas pressures.

Discussion

In the experiments, we used sufficiently monodisperse spherical lycopodium particles (fern spores). Their projective diameters fall in the range from 20 to 32 µm, and the density (according to the reference data) varies insignificantly: $\rho_p = 0.95 - 1.24 \ g/cm^3$. The lycopodium powder is hygroscopic; therefore, before the experiment the particles were subjected to vacuum drying, after which their density was estimated as $1.0 \ g/cm^3$. The lycopodium particles are rather well subject to electric charging in the corona discharge in an ionizer and carry $(2-5)\cdot 10^5$ elementary electric charges on the surface.³ The accurate size of the lycopodium particles was determined just in the experiment also by the stability loss method.^{1,3}

Figure 3 shows, in the standard form, the experimental results on the reduced drag force F_U^* for a lycopodium particle in air and the theoretical predictions as functions of the Knudsen number Kn. Here

$$F_U^* = F_U(\mathrm{Kn}) / F_U^{\mathrm{im}}, \qquad (2)$$

where the free-molecule value of the drag force at complete accommodation of the momentum and energy of gas molecules on the particle surface⁵ is determined as

$$F_U^{\rm fm} = \frac{16\sqrt{\pi}}{3} \left(1 + \frac{\pi}{8}\right) R_{\rm p}^2 p_\infty \sqrt{\frac{M}{2RT_\infty}} U_\infty \,, \qquad (3)$$

where M is the gas molar mass; R is the absolute gas constant; p_{∞} is the gas pressure; T_{∞} is the gas temperature; U_{∞} is the rate of the incident gas flow; the subscript ∞ corresponds to the gas state at a sufficient distance from the particle. The formula approximating the numerical calculations with the error no greater than 1% in the whole range of Knudsen numbers Kn (curves 1-3) has the form⁵:

$$F_U^*(\mathrm{Kn}) = \frac{\mathrm{Kn}}{\mathrm{Kn} + 0.619} \left(1 + \frac{0.310\mathrm{Kn}}{\mathrm{Kn}^2 + 1.152\mathrm{Kn} + 0.785} \right) \times \frac{1 - (1 - \alpha_\tau) \left(1 - \frac{0.641\mathrm{Kn}}{\mathrm{Kn} + 0.158} \right)}{1 - (1 - \alpha_\tau) \left(1 - \frac{\mathrm{Kn}}{\mathrm{Kn} + 0.168} \right)},$$
(4)

where α_{τ} is the tangential momentum accommodation coefficient, whose known values for the so-called engineering surfaces (including aerosols) lie in the range from 0.8 to 1.0. The experimental values of the drag force were determined using both numerical solution for the stability threshold curve¹ and the approximate solution¹¹ in order to compare the results obtained.



Fig. 3. Reduced drag force F_U^* for lycopodium particle with $R_p = 13.9 \ \mu\text{m}$ in air vs Knudsen number: experiment with calculation of the stability threshold by the finite difference method¹ (dots); experiment with approximate calculation of the stability threshold by the results from Ref. 11 (crosses); theory⁵ at $\alpha_{\tau} = 1.0$ (1), 0.9 (2) and 0.8 (3).

At small values of the Knudsen number Kn $\approx 0.1-0.2$, the experimental data systematically insignificantly exceed the theoretical predictions. This fact can be explained by small, though finite value of the Reynolds number Re at particle oscillations about the equilibrium position with the gas pressure close to the atmospheric one. The estimates by the Oseen formula show that this inertial effect is marked already at Re ≈ 0.1 . The theoretical results for $F_U(\text{Kn})$ are traditionally analyzed under condition Re $\rightarrow 0$ (Refs. 4 and 5). At the intermediate values of the Knudsen number Kn $\approx 0.2-1.0$, the experiment and the theory are in a very close agreement at $\alpha_{\tau} = 0.9-1.0$; the inertial effect here is already insignificant.

Indeed, the Reynolds number is defined as Re = $= 2\rho U_{\infty}R_{\rm p}/\eta$. As the pressure decreases, the gas density ρ decreases too, while the viscosity coefficient η is pressure independent, which leads to a decrease in the realized Reynolds numbers Re. At larger Knudsen numbers Kn and further decrease of the gas pressure, the Brownian motion of the particle intensifies, and this significantly hampers the measurements and decreases their accuracy. This problem is of principal importance in the experimental investigations of various phenomena at $Kn \ge 5$, limiting the possible range of modeling the processes with atmospheric aerosol at high altitudes and requiring the increase of statistical confidence of the results obtained (increase of the number of measurements of stability loss events at the fixed gas pressure).

The observed decrease of the experimental values of the drag force at moderate and large values of the Knudsen number Kn, as compared to the theory, is not likely connected with the known effect of space constraint. Indeed, the Knudsen number estimated by the characteristic size of the measuring cell $\text{Kn}_H = l/2H$, where 2H is the separation between the disc electrodes (see Fig. 1), even at the limiting degrees of chamber evacuation is rather small ($\text{Kn}_H \le 0.01$). Therefore, the approximation of a particle in an infinite gas volume that was accepted in Refs. 4 and 5 is always valid within the accuracy needed.

It is also interesting to evaluate how the charge of an aerosol particle affects the drag force measured by the EDL method. In theoretical description of both the drag force and other phoretic effects, aerosol particles were traditionally assumed uncharged (we do not know other results).

The unexpectedly strong effect of the sign and density of the charge of various microparticles in different gases on the thermophoretic force was noticed in Ref. 12. This new effect is especially significant for helium and less marked for air. The experiments in Ref. 12 were conducted by the EDL method with the double ring electrodes in a vacuum chamber at variable gas pressure. Li and Davis¹² explain the effect discovered by the electrostatic interaction between the charged particle and molecules of nonpolar gases used in the experiment. The polarizability of the likely changes rather molecules strongly the characteristics of their interaction with the charged surface of the particle. Thermophoresis of particles is associated with energy transfer in a gas and described by such a transfer characteristic as the heat conductivity. The drag force is caused by momentum transfer in a gas and characterized by the viscosity coefficient, which also can be sensitive to the polarization effects.¹³

Comparison of the obtained data with the theory^{4,5} and the known theoretical results obtained by other authors for uncharged aerosol particles does not reveal any marked effect of the particle charge on the drag force within the measurement accuracy. The same conclusion follows also from analysis of the well-known Millikan experimental data (see, for example, Ref. 2). In these experiments, particles were also subjected to charging in order to hold them in an electrostatic levitator. This issue calls for further analysis and study, since the phenomenon discovered in Ref. 12 is not connected with the effect of known thermomagnetic forces (see, for example, Ref. 14) explained by the Beenakker-Senftleben effect.¹³

Conclusion

In this paper, the drag force of lycopodium particles in air has been measured by the EDL method at the Knudsen number varying in a rather wide range Kn = 0.1-3.0. It has been shown that in

the experiment there is no need to generate specially the incident air flow, since the relative motion of the particle and the gas at low values of the Reynolds number is realized due to oscillations of the particle as it leaves the stability threshold curve. Comparison of the experimental data with the molecular-kinetic theory of the studied phenomenon^{4,5} shows quite close agreement between them in the whole range of the Knudsen number realized in the studies with the molecular tangential momentum accommodation coefficient $\alpha_{\tau} = 0.9-1.0$.

The experiments conducted are also considered as test dynamic measurements carried out by the EDL method for the particular experimental setup. They confirm the efficiency and adequacy of the technique developed to the requirements of modeling the processes with atmospheric aerosol at high altitudes. In the future, this technique will be used for experimental investigation of photophoresis of stratospheric aerosols in the field of solar and outgoing thermal Earth's radiation.¹⁵

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