Experimental study of thermophoresis of aerosol particles under microgravity conditions

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Experimental estimations of thermophoretic velocity of spherical copper particles in nitrogen measured in microgravity conditions at the Bremen drop tower (Germany) at very low values of the Knudsen number (Kn ≈ 0.002) and very high thermal-conductivity parameter Λ ≈ 20 500 are presented. Comparison of the obtained data with the gas-kinetic theory, taking into account both thermal-creep flow and thermal-stress slip flow mechanisms of thermophoresis is carried out. The obtained results demonstrate indirectly the action of the thermal stress slip flow mechanism, however, negative thermophoretic velocities have not been recorded. The important role of energy accommodation coefficient for gas molecules on the particle surface for explanation of the obtained results is discussed. An additional analysis of experimental data is necessary, aiming at the increase of reliability of thermophoretic velocity estimates.

Introduction

Thermophoresis of aerosols (directed motion of particles in the field of the temperature gradient) is the subject of many-year experimental and theoretical investigations because of great significance of this phenomenon for basic and applied aerosol microphysics.[1] In spite of relatively small magnitude of the characteristic temperature gradients in the stratosphere as compared to that used in different technological processes, thermophoresis can play an important role in cloud physics and cleaning the atmosphere from aerosol particles.[2]

Let us briefly recall the mechanisms of appearance of thermophoretic force at small Knudsen numbers. If thermal conductivity of a particle is relatively small, then its side, facing the "hot" gas area becomes more heated, and the induced temperature gradient appears on the particle surface, comparable with analogous gradient in the gas. As the result, thermal slip of the gas (thermocreep), directed from cold area to hot one appears along the surface, and the particle undergoes the force in the direction opposite to the temperature gradient in the gas (the first, traditional mechanism of appearance of thermophoretic force at small Knudsen numbers[3]). When thermal conductivity of the particle significantly exceeds the thermal conductivity of the gas, the surface temperature becomes practically homogeneous, and the effect of thermocreep mechanism almost disappears. However, the tangent component of the thermal tension on the particle surface does not become zero, and the thermal slip of the second order in Knudsen number appears (so called thermal-stress gas slip) directed from the hot area to the cold one. As the result, the particle should move along the temperature gradient in the gas (so called negative thermophoresis, associated with the second mechanism of the force appearance at small Knudsen numbers[4,5]).

Negative thermophoresis (motion of aerosol particles to the hot area of the gas) was predicted earlier in a number of works, but estimates of thermophoretic velocity and the ranges of manifestation of the effect in Knudsen numbers are different in different theoretical models. General necessary conditions are small magnitude of the Knudsen number $\mathrm{Kn} = l/R_p \ll 1$ (where l is the mean free path length of gas molecules, R_p is the particle radius) and high thermal conductivity of the particle with respect to the gas $\Lambda = \lambda_p/\lambda_g \gg 1$ (where λ_p and λ_g are the thermal conductivity of the particle and transmission thermal conductivity of the gas, respectively).

To date, the negative thermophoresis has been not experimentally observed, in spite of numerous attempts. The only indirect confirmation of the effect was discussed in Ref. 6 (see also comments in Ref. 1). The microgravity conditions are the most suitable, if any, for experiments on observing the negative thermophoresis. This was noted sufficiently long time ago.[7] The distorting effect of gravity and gravitational convection on the fields of density, temperature, and the velocity of motion of the gas near the particle are excluded from them to some degree. This method was successfully proved in investigations of thermophoresis of particles of moderate thermal conductivity, which enabled one to quantitatively compare the experiment and the theory.[8]

This paper presents experimental estimates of thermophoretic velocity of spherical copper particles in nitrogen, measured under microgravity conditions at the Bremen drop tower (Germany) at extremely

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small values of the Knudsen number (Kn ≈ 0.002) and very high values of thermo-physical parameter Λ ≈ 20 500. The experimental results were preliminary compared with the gas-kinetic theory taking into account not only thermo-creep but also thermo-stress mechanisms of appearance of the force at small Knudsen numbers.

Theory

For adequate comparison with the experiment, it is necessary to use for the above range of the determining parameters the theory of thermophoresis, which correctly (quantitatively and qualitatively) takes into account the mechanisms of appearance of the force at small Knudsen numbers. Sparse results obtained based on the solution of the linear Boltzmann equation with potential of intermolecular interaction for the model of solid sphere are most reliable today.[5,9–11] Unfortunately, only thermophoretic force has been used in all calculations, while the data on the thermophoretic velocity are required for comparison with the conducted experiment. Transition to the thermophoresis velocity in this case is not a trivial problem, because it requires the knowledge of the resistance force, calculated at the same level of accuracy in the Knudsen number.

Besides, the model of purely diffuse scattering was applied in Refs. 9–11 for the distribution function of reflected molecules, which corresponded to the case of full accommodation of all molecular features on the particle surface. The Maxwell mirror-diffuse scheme of boundary conditions[5] (for three particular values of the mirror reflection coefficient) also is not sufficiently adequate for the considered problem, as it will be shown below.

Therefore, the thermophoresis theory results, obtained on the basis of solution of the linearized S-model gas-kinetic equation,[12] were chosen for comparison with experimental data. It is known that this model equation correctly describes the combined processes of energy and pulse transfer in the one-atom gas (as opposite to the well-known BGK-model[13]) and provides for true values of all 13 moments for the gas molecule velocity distribution function associated with macroscopic velocity, temperature, tensor of tensions, and heat flux in the gas. In general, the problem of a priori accuracy of the model gas-kinetic equations of a sufficiently high order in comparison with the complete linearized Boltzmann equation remains open; usually it is solved through comparison of the obtained results for the known test problems. Such detailed comparison was not carried out to date for the problem of thermophoresis, however, the analysis of results for the problems on isothermal and thermal gas slip has demonstrated a high a priori accuracy of the S-model kinetic equation and the possibility of its successful application to the problems of aerosol mehcanics.[14]

The possibility of arbitrary values of the thermo-physical parameter Λ, as well as accommodation coefficients of energy α_E and tangent pulse α_τ of gas molecules on the particle surface is presumed in Ref. 12, initially oriented to the practical use of the obtained results. Both the force and the velocity of thermophoresis are calculated there in the total range of Kn numbers, therewith, asymptotic formulas are obtained for the range Kn ≪ 1, taking into account the terms of the order $O(Kn^2)$. The error, accepted for calculations by the Bubnov–Galerkin method, does not exceed 2% at Kn ≈ 0.01; the maximal error in calculations is estimated as 0.5% at any values of the varying parameters.

Experiment

For appearance of the thermophoretic force in experiments, it was necessary to gain the particle motions under the prevalence of the mechanism of thermal-stress gas slip. To do this, it was necessary to realize very small Kn number values and very high values of the thermal-physical parameter Λ. The requirement of the number Kn smallness forces one to use the large-size particles and quite high pressures of the surrounding gas. In this work, the Kn values of about 0.002 and Λ ≈ 20 500 were reached, when dispersing spherical copper particles with a mean diameter of 74 μm in nitrogen under normal pressure.

In the mode of prevalent thermal-stress gas slip, the thermophoretic velocities of particles are extremely small in comparison with the velocities caused by other forces. For example, estimates of thermophoretic velocities under the above conditions and a temperature gradient of $5 \cdot 10^4$ K/m provide for the range from –5 to +20 μm/s depending on the values of the accommodation coefficients of the gas molecule energy, while the velocity of gravitational sedimentation of these particles under the same conditions is close to 1 m/s. The inevitably appearing gravitational convection at such temperature gradients leads to intensive gas motion, which technically is hardly suppressible to the values, comparable with thermophoretic velocities, because of disturbances introduced into the thermal field of the measuring cell by the presence of the systems of particle injection and observation (windows, light sources), as well as systems of gas inlet-outlet and ionization.

In order to suppress particle sedimentation and gravitational convection, the experiments were conducted under conditions of free fall in a vacuum tube of a height of 120 m of the University of Bremen drop tower (Germany), which provided for microgravity of high quality (residual acceleration less than 10^{-5} g) during 4.7 s. The equipment was located in the experimental section of a hermetic capsule with an inner diameter of 0.6 m and a height of 1.7 m. The total mass of the capsule was about 500 kg. A standard pressure and room temperature were maintained in the capsule.

When planning the experiments, it was necessary to take into account that the studied system before dropping was under standard gravity conditions, then the conditions very fast changed to microgravity with duration of 4.7 s, which ended by an overload of up to 50 g, when braking the capsule with the equipment.

Relatively short time of microgravity conditions imposed restrictions on the cell size. Depending on geometry, its inner volume should not exceed 1–2 cm^3 in order the relaxation times of the processes of energy and pulse transfer do not exceed ~ 0.5 s, which is significantly less than the total duration of the microgravity conditions. Thus, the convective motion and thermal fields, related with gravitational convection before dropping, quickly damp, and stationary conditions characteristic of free fall, appear and remain in the measuring cell for 4 s.

Before dropping, spherical copper particles were placed on the nylon net at the bottom of the cell (*1*) (Fig. 1).

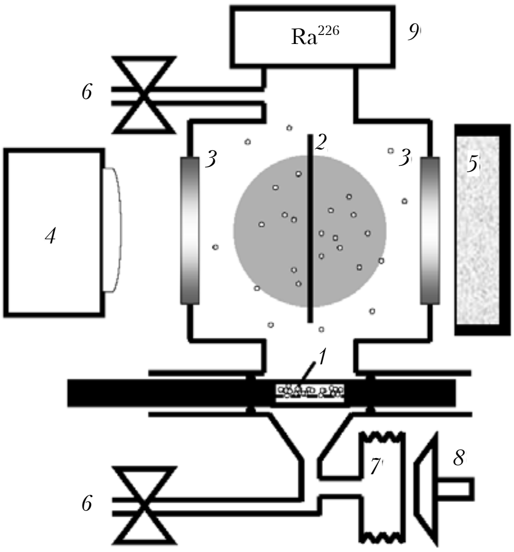

Fig. 1. Schematic view of the experimental setup: particles on nylon net before injection into cell (*1*); heated thread (*2*); four windows (*3*); one of two digital video-cameras (*4*); background illumination (*5*); tubes with valves for inlet and outlet of the working gas (*6*); membrane (*7*); percussive mechanism for injecting particles (*8*); source of gas ionization (*9*).

The cell was filled with nitrogen by means of two valves *2*. The temperature gradient was created by transmitting the current through a metal thread *3* of 300 μm in diameter in the cell center (Figs. 1 and 2) in parallel to windows and side walls of the cell. The heated thread was fastened to two ceramic tube-holders of 1.5 mm in diameter. The temperature was controlled by thin thermocouples, one of which touched the thread. The field and the temperature gradients were calculated based on the known temperatures of the thread (about 120°C), walls (30°C), and the control point inside the cell.

Just at the moment of establishing the microgravity conditions, copper particles were injected into the experimental cell by means of single brief and intensive forward-reverse gas motion near the net with lying particles. The particles were scattered over the cell volume, mainly mechanically. The above mode was provided for by the membrane *8* and the percussive mechanism *9* consisting of the electromagnet with mobile core and returning spring. The short-time current traversed through the electromagnet provided for one blow on the membrane followed by the return to the initial position. Duration of the injection was quite short (less than 1 μs), it did not change the temperature distribution in the cell and did not create convective gas flow. Analysis of trajectories of the particle motion after injecting has confirmed the relaxation time to correspond to initial estimates.

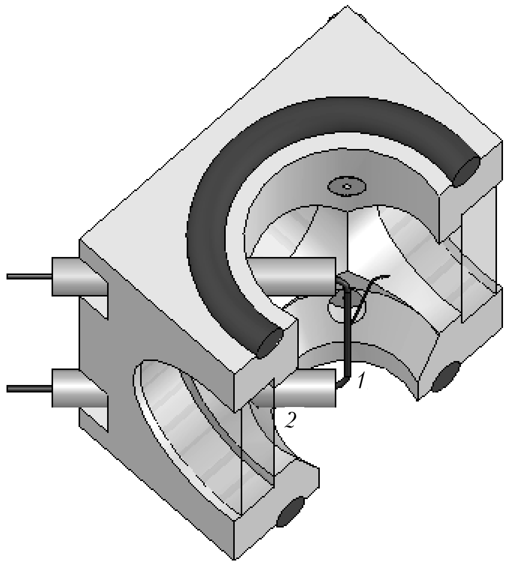

Fig. 2. Vertical section of the measurement cell for studying thermophoresis under microgravity conditions: heated thread (*1*); ceramic holders (*2*); window (*3*).

The cell had four windows *3* arranged at right angles for stereoscopic observation of particles. Pairs of stereoscopic images in the cell center around the heating thread were recorded with frequencies of 15 and 30 frame/s. The values of temperature and temperature gradients were assigned to every particle according to its calculated distance to the thread. To decrease the possible effect of the particle electrization, the source of α-radiation (Ra^{226}) *7* was used. In order to estimate disturbances, not related to the heated thread (residual microaccelerations, electrostatics, etc.), test experiments were carried out under microgravity conditions in the absence of the temperature gradient.

Discussion

Only radial components of particle velocities in the experiment can carry information about their thermophoretic motion, because the field of temperature gradient near the heated thread, where the particle motion is recorded, is determined by the cylindrical symmetry of the problem (see Fig. 2).

Other components of the particle motion velocities can be used for estimation of possible configurations of appearing gas flows in the measurement cell. The observed radial motion of particles was quite stable in time and direction at quite wide scatter of the velocity values. The latter did not obey to the inverse proportional dependence on the radial coordinate, which should be observed in an ideal situation of purely thermophoretic motion of particles in the temperature field of the heated thread. Authors ascribe these deviations to the influence of non-gravity convection on particles, caused by the gas thermal slip (thermocreep) along construction elements of the cell, which cause large temperature gradients of the heated thread itself and its ceramic holders.

Numerical simulation of the gas velocity field in the measurement cell

The measurements cell was simulated as a cylinder, the height and diameter of which were the same as of the actual cell, but windows were neglected. The patterns of gas motion velocities in the plane crossing the thread holders (to the left) and in the equator plane perpendicular to the heated thread (to the right) are shown in Fig. 3.

The calculated velocity field has a complicated configuration and is characterized both by radial (the most important for analysis) and non-radial components of the velocity. High non-radial velocities, caused by the gas thermal slip, are characteristic of the cell areas, where the holders were placed (see Fig. 2); these velocities are much less on the opposite side from the holders. The effect of the holders on the velocity field is minimal in the cell center. Non-radial velocities in the equator plane are zero near the thread and quickly increase at deviation from this plane. The radial component of the velocity tends to zero when approaching to the thread and increases at moving away from the center, taking values of about 10–20 μm/s at distances of 2–3 mm, where particles were mainly recorded. Particular values of gas velocities also depend on position of the observation point relative to the holders.

Estimation of thermophoretic velocity of particles

The analysis of the velocity field has shown that particles moving just near the thread in the cell center provide for the most reliable data for estimation of thermophoretic velocities. The experimentally measured velocities were corrected taking into account the data of numerical simulation. They are shown in Fig. 4 as the dimensionless reduced thermophoretic velocity

$$V_{\mathrm{thr}} = -V_{\mathrm{th}} T / (\nu \nabla T),$$

where V_{th} is the measured velocity of the particle motion; T and ∇T are the temperature and the temperature gradient near the particle; ν is the kinematic viscosity of the gas. The corrected thermophoretic velocities V_{thr} are shown in Fig. 4 in two variants.

In the first variant they are averaged for all particles undergone radial thermophoretic motion in the whole measurement volume, with the same statistical weight. They gave the most conservative estimate of the reduced thermophoretic velocity: $V_{\mathrm{thr}} = 0.022 \pm 0.038$. This value is, in the authors' opinion, the upper boundary of the experimental estimate of V_{thr}. Thermophoretic velocity is positive; however, characterized by very great confidence interval.

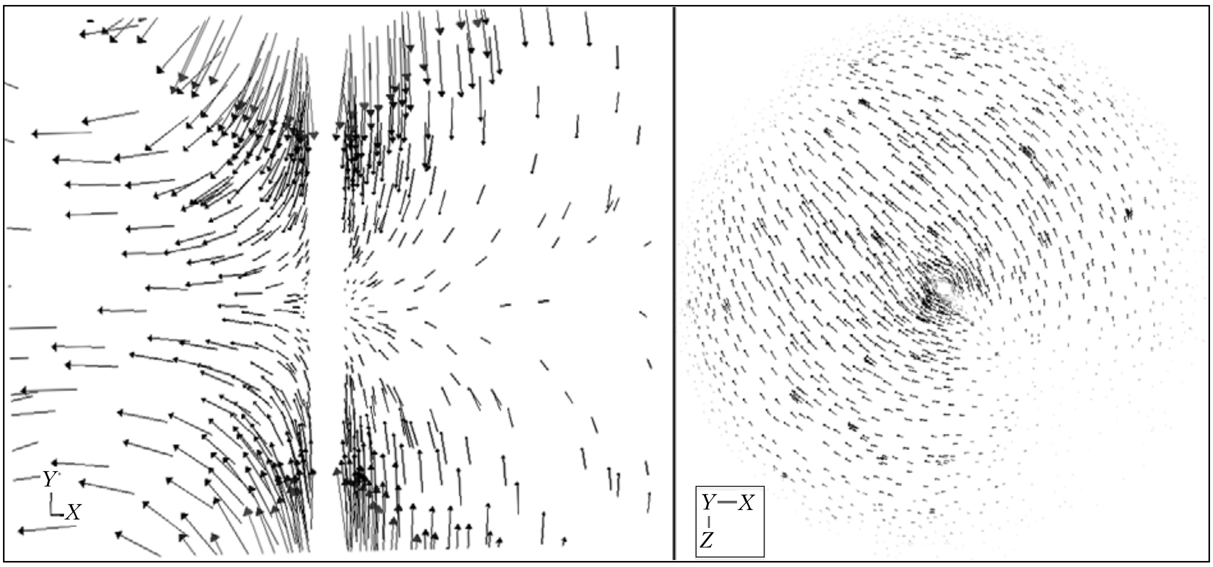

Fig. 3. The field of the gas motion velocity, caused by thermal slip on nonhomogeneously heated thread holders and on the thread itself. To the left, the velocity field plane passes through the holders (they are inside the visual zone); to the right, the velocity field plane is perpendicular to the thread at its middle point.

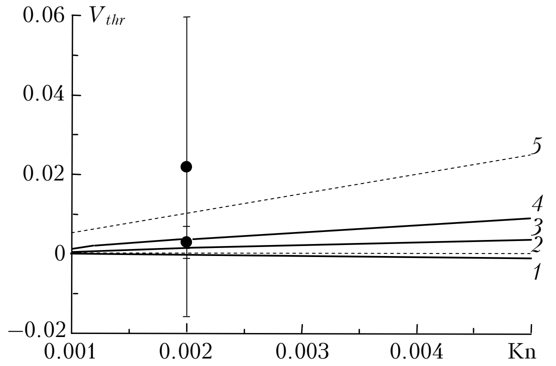

Fig. 4. Comparison of experimental and theoretical dimensionless thermophoretic velocities V_{thr} of copper particles in nitrogen ($\Lambda \approx 20\,500$): theoretical results[12] at accommodation coefficient of the tangent pulse $\alpha_\tau = 1.0$ and the energy accommodation coefficient $\alpha_E = 1.0$ (*1*); at 1.0 and 0.95 (*2*); at 1.0 and 0.9 (*3*); at 1.0 and 0.80 (*4*), respectively; hydrodynamic theory of thermophoresis[3] (*5*). Upper black circle shows the mean (conservative) experimental estimate of V_{thr} with the confidence interval; lower circle shows the "optimistic" experimental estimate of V_{thr}.

In the second variant, from all particles undergone radial motion, those particles were selected, which were located close to the thread in the cell center, thus undergoing less influence of the disturbing factors. Assigning to them greater weights at the averaging, we obtain the "optimistic" estimate of the reduced thermophoretic velocity: $V_{\text{thr}} = 0.003 \pm 0.004$.

Comparison of experiment and theory

In addition to experimental data, the thermophoretic velocities, calculated at small Kn numbers by the theory[12] are shown in Fig. 4. In the calculations, the values of accommodation coefficients of gas molecules on the particle surface are set along with the thermo-physical parameter $\Lambda \approx 20\,500$: tangent pulse $\alpha_\tau = 1.0$ and energy $\alpha_E = 0.80 \div 1.0$. The used values are well warranted (see, for example, Ref. 15) and correspond to the case of so called "technical" (covered by a layer of adsorbent) copper surface in nitrogen. For the atomic-clean surface (which is unreachable in the considered experiments) lower α_E values can be expected.[15]

Ideally, when comparing theory and experiment, not the interval of possible values of the accommodation coefficients should be set, but their particular values, determined from independent experiments with the same particles and gases. Such point of view is not new, it is regularly discussed,[16,17] but is not yet realized in practice.

Theoretical values of the reduced thermophoretic velocity for $\alpha_E = 1.0 \div 0.975$ (not shown in Fig. 4) are negative, and become positive at less values of α_E. It is well known that the force and velocity of thermophoresis are very sensitive to variations of α_E (both at thermocreep and thermostress appearance of the effect[5,18]). Evidently, its value governs the magnitude of the heat flux, normal to the surface, which, in its turn, controls the competition of oppositely directed thermocreep and thermostress gas flows near the particle surface.

Thus, the attempts of revealing negative thermophoresis lead to unexpected conclusion: the force and velocity of thermophoresis can be positive even for strongly thermal-conductive particles at very small Kn numbers, because this is determined by a particular α_E value for a particular "gas–particle" system. Note that the known theoretic predictions of negative thermophoresis[9–11] were made in assumption of completely diffuse scattering of gas molecules on the particle surface, that corresponds to the obligatory condition: $\alpha_E = 1.0$.

The curve *5* in Fig. 4 corresponds to the known "direct hydrodynamic" theory of thermophoresis[3] for the case $\alpha_\tau = \alpha_E = 1.0$ recommended for practical use, when particles have moderate thermal conductivity at small Knudsen numbers. By the method of its construction, it takes into account only thermocreep (but not thermostress) mechanism of thermophoresis. It is seen in Fig. 4 that the "optimistic" experimental estimate of the reduced thermophoretic velocity agrees much better with the results of gas-kinetic theory[12] at quite realistic and permissible values of α_E, than with predictions of the "hydrodynamic" theory.[3]

Conclusions

Thus, the conducted experiments quite convincingly have demonstrated indirect manifestation of the mechanism of the thermophoresis phenomenon appearance for strongly thermo-conductive particles at very small Knudsen numbers; however, negative thermophoretic velocities were not recorded. Evidently, an additional analysis of experimental data is necessary in order to increase the unambiguity of the "optimistic" estimate of the thermophoretic velocity.

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