Thermophoretic interaction of heat releasing particles

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This study investigates thermophoretic force acting at heat releasing (absorbing) particles near the interface between two media with different thermal conductivities. This force is caused by the induced temperature gradient which is proportional to the rate of heat release (absorption) by the particle. Therefore the magnitude of the thermophoretic force is proportional to the rate of heat release (absorption) by the particle, and its direction depends upon the sign of the parameter $\kappa_1 - \kappa_2$, where κ_1 is thermal conductivity of a host medium and κ_2 is thermal conductivity of the adjacent medium. The obtained results imply that a heat releasing (absorbing) particle is attracted (repelled) to the interface when thermal conductivity of a host medium is less than thermal conductivity of the adjacent medium. Thus, e.g., growing in air by condensation particle is attracted to a metal surface while an evaporating in air particle is repelled from a metal surface. The change of temperature distribution caused by heat releasing particles results in the additional thermophoretic interaction of these particles. We determined a condition for mutual attraction of two spherical heat releasing particles and derived an expression for the thermophoretic force acting at the particles. The magnitudes of the considered thermophoretic forces are compared with the classic thermophoretic force. [DOI: 10.1063/1.1556196]

I. INTRODUCTION

Behavior of microparticles inside a host medium attracted considerable attention because of the emergence of new technological processes involving microparticles, their significance in atmospheric physics and due to a large variety of mechanisms determining the behavior of microparticles (see Refs. 1–3 and references therein). One of the mechanisms determining the behavior of small particles in fluids is associated with thermophoretic forces. These forces arise due to the kinetic slip of a fluid flow near the particle in the presence of the temperature gradient along its surface.[3,4]

When temperature gradient is caused by an external source then the magnitude of the effect depends upon thermal conductivity of a host medium κ_1 and of the particle κ_p. In the limiting case when $\kappa_p \to \infty$ this effect disappears. Different situation occurs when a particle is located near the boundary between two media and when the particle itself is a heat source or sink. The latter can occur, e.g., in the case of a chemically active particle when heat is released or absorbed due to a chemical reaction. In this situation thermal conductivity of a particle is of lesser importance since the induced at the particle temperature gradient due to the presence of the interface between two media is determined primarily by thermal characteristics of the host and adjacent media.

In Sec. II of this study we investigated a force acting at the particle due to the induced temperature gradient caused by the presence of the interface between two media with different thermal conductivities. Although the effect of thermophoresis was discovered more than one hundred years ago (see, e.g., Ref. 4 and references therein) the feasibility of the thermophoretic self-action of a heat releasing particle near the interface was not discussed before. The obtained results imply that a heat releasing (absorbing) particle is attracted (repelled) to the interface when thermal conductivity of a host medium is less than thermal conductivity of the adjacent medium. Thus, e.g., growing in air by condensation particle is attracted to a metal surface while an evaporating in air particle is repelled from a metal surface.

Using the results obtained in Sec. II of this study in Sec. III we determined a condition for mutual attraction of heat releasing particles and derived an expression for the thermophoretic force acting at the particle.

II. FORCES ACTING AT A HEAT RELEASING PARTICLE LOCATED NEAR THE BOUNDARY BETWEEN TWO MEDIA WITH DIFFERENT THERMAL CONDUCTIVITIES

Consider a plane surface $z=0$ separating two media with thermal conductivities κ_1 and κ_2, correspondingly. Assume that a spherical heat releasing particle with a center at $\mathbf{c}$ having a radius a and a heat release rate q is located inside a host fluid with thermal conductivity κ_1 (see Fig. 1). The host fluid is considered to be incompressible and Newtonian, and we neglect temperature dependence of the physical properties. Assume small Knudsen and Reynolds numbers, $\mathrm{Kn} \ll 1$ and $\mathrm{Re} \ll 1$, so that inertial effects can be neglected.

The force acting at a particle in a stationary regime is determined by the following system of equations:

$$\kappa \nabla^2 T + q = 0, \quad \nabla p = \eta \nabla^2 \mathbf{u}, \quad \nabla \cdot \mathbf{u} = 0. \tag{1}$$

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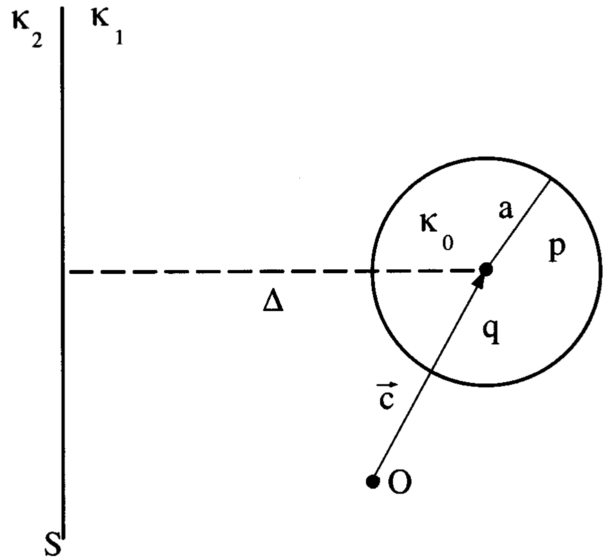

FIG. 1. Location of a particle with a radius a near the interface between two media with thermal conductivities κ_1 and κ_2.

Here T is temperature, $\mathbf{u}$ is a velocity of a host fluid with an imbedded heat source (particle) q, p is fluid pressure, and η is a dynamic viscosity. Equation (1) must be supplemented with boundary conditions for temperature and velocity. When velocity $\mathbf{u}$ is found then the force acting at the particle is determined as follows (see, e.g., Refs. 1 and 5):

$$\mathbf{f}=\int \hat{\sigma}\cdot d\mathbf{S}, \quad \hat{\sigma}=-p\hat{e}+\eta(\nabla\mathbf{u}+(\nabla\mathbf{u})^T), \tag{2}$$

where $\hat{\sigma}$ is a stress tensor, and $\hat{e}$ is a unit tensor and integration is performed over the surface of a particle. Boundary conditions for temperature read

$$[T]_S=0, \quad \mathbf{n}\cdot[\kappa\nabla T]_S=0, \tag{3}$$

where $\mathbf{n}$ is a unit external normal vector, and $[\beta]_S=\beta_S^+-\beta_S^-$, $\beta_S^\pm$ are values of β at the external and internal surfaces separating two media, respectively. Hereafter subscript S denotes that a corresponding variable is determined at the surface separating between two media, while subscript p implies that variable is determined at the surface of the rigid particle. Boundary conditions for the velocity are as follows:

$$\mathbf{u}_\infty=0, \quad \mathbf{u}_S=0 \quad (\mathbf{n}\cdot\mathbf{u})_p=0,$$
$$(\boldsymbol{\tau}\cdot\mathbf{u})_p=\vartheta(\boldsymbol{\tau}\cdot\nabla T)_p, \tag{4}$$

where $\boldsymbol{\tau}$ is a unit tangential vector, $(\ )_\infty$ denotes the magnitude far from the particle, and $(\ )_p$ denotes the value at the particle's surface. A coefficient ϑ in Eq. (4) is considered to be constant in order to linearize the problem, and its dependence upon the parameters of the problem is discussed below.

The first condition in the boundary conditions (4) implies that a particle is at rest in the laboratory frame of reference. Thus, it is assumed that an external force is applied at the particle in order to compensate a force determined by Eq. (2). The second condition in the boundary conditions (4) implies the absence of slip at the surface separating between the fluid and an external solid medium. Slip may occur due to the temperature gradient induced by a heat source when a heat source is located near the edges of this surface. Here we consider a situation when a heat source is located far from the edges and the surface can be considered as an infinite. The third boundary condition in Eq. (4) corresponds to a rigid particle, and the last condition in Eqs. (4) implies a slip of fluid at the particle due to the induced by the surface temperature gradient.

Hereafter we assume that a heat releasing particle is located in the region $z>0$. The Green's function $G(\mathbf{r},\mathbf{r}')$ which satisfies an equation

$$\nabla^2 G(\mathbf{r},\mathbf{r}')=\delta(\mathbf{r}-\mathbf{r}')$$

and boundary conditions (3) with respect to variable $\mathbf{r}$ is well known (see, e.g., Ref. 6, Chap. 2, Sec. 7). Under the condition that a point $\mathbf{r}$ is located in a region 1, $G(\mathbf{r},\mathbf{r}')\equiv G_1(\mathbf{r},\mathbf{r}')$ and

$$G_1(\mathbf{r},\mathbf{r}')=-\frac{1}{4\pi}\left(\frac{1}{|\mathbf{r}-\mathbf{r}'|}+\frac{\kappa_1-\kappa_2}{\kappa_1+\kappa_2}\frac{1}{|\mathbf{r}-\mathcal{R}\mathbf{r}'|}\right). \tag{5}$$

Here $\mathcal{R}$ is an operator of specular reflection with respect to a plane $z=0$, i.e., $\mathcal{R}\mathbf{r}=(x,y,-z)$.

If a point $\mathbf{r}$ is located in region 2, then $G(\mathbf{r},\mathbf{r}')\equiv G_2(\mathbf{r},\mathbf{r}')$, and

$$G_2(\mathbf{r},\mathbf{r}')=-\frac{1}{4\pi}\frac{2\kappa_1}{\kappa_1+\kappa_2}\frac{1}{|\mathbf{r}-\mathbf{r}'|}. \tag{6}$$

Then the solution for the temperature reads

$$T(\mathbf{r})=\frac{1}{\kappa_1}\int G(\mathbf{r},\mathbf{r}')q(\mathbf{r}')d\mathbf{r}'. \tag{7}$$

Expression (5) for the Green function implies that temperature distribution $T(\mathbf{r})$ is given by a sum of two terms. The first term describes temperature distribution due to the heat source, and the second term is associated with the presence of the interface. The latter term is responsible for the effect of self induced thermophoresis, while the contribution of the first term is zero due to boundary conditions (4). The formula for the second term reads

$$T_1(\mathbf{r})=\frac{1}{\kappa_1}\frac{\kappa_1-\kappa_2}{\kappa_1+\kappa_2}\frac{1}{4\pi}\sum_{l,m}\frac{Y_{lm}(\mathbf{n})\rho_{lm}}{|\mathbf{r}-\mathcal{R}\mathbf{c}|^{l+1}}, \tag{8}$$

where

$$\rho_{lm}=\frac{4\pi}{2l+1}\int_0^a\int_\Omega Y_{lm}^*(\mathcal{R}\mathbf{n}')a'^{l+2}q(a',\mathbf{n}')da'\,d\Omega(\mathbf{n}'),$$

$$\mathbf{n}=\frac{\mathbf{r}-\mathcal{R}\mathbf{c}}{|\mathbf{r}-\mathcal{R}\mathbf{c}|}, \quad \mathbf{n}'=\frac{\mathbf{r}'-\mathbf{c}}{|\mathbf{r}'-\mathbf{c}|},$$

$a'=|\mathbf{r}'-\mathbf{c}|$, and $Y_{lm}(\mathbf{n})$ are spherical functions (see, e.g., Ref. 7).

If coefficients ρ_{lm} have azimuthal symmetry with respect to the axis connecting two points $\mathbf{c}$ and $\mathcal{R}\mathbf{c}$, then T_1 can be written as follows:

$$T_1(\mathbf{r})=\frac{\xi_\kappa}{2\kappa_1}\sum_{l=0}^\infty\frac{P_l(\mathbf{n})\rho_l}{|\mathbf{r}-\mathcal{R}\mathbf{c}|^{l+1}}, \quad \xi_\kappa=\frac{\kappa_1-\kappa_2}{\kappa_1+\kappa_2},$$

$$\rho_l=(-1)^l\int_{-1}^{1}\int_{0}^{a}P_l(\mu')q(a',\mu')a'^{l+2}da'\,d\mu',$$

and $P_l(\mu)$ are Legendre polynomials.[7]

Keeping only the first nonvanishing terms with respect to a parameter a/Δ, where Δ is a distance between the center of the particle and the interface between the two media, we obtain the following expression for $\boldsymbol{\nabla}T_1$:

$$\boldsymbol{\nabla}T_1=-\frac{\xi_\kappa\mathbf{k}\rho_0}{2\kappa_1(2\Delta)^2}, \tag{9}$$

where

$$\mathbf{k}=\frac{\mathbf{c}-\mathcal{R}\mathbf{c}}{|\mathbf{c}-\mathcal{R}\mathbf{c}|}.$$

Thus, in the first nonvanishing order with respect to the parameter a/Δ, the induced temperature gradient field is homogeneous, and its magnitude is determined by the geometry of a heat source q.

In order to solve a hydrodynamic part of the problem one can use either bispherical coordinates[8] or employ a method of reflections (see Ref. 2, p. 189). In the latter case a velocity field is determined by the following expansion:

$$\mathbf{u}=\mathbf{u}_0+\mathbf{u}_1+\mathbf{u}_2+\ldots,$$

where each term is determined by boundary conditions (4) and a boundary condition which is satisfied by a preceding term. Thus if $\mathbf{u}_0$ is determined by boundary conditions (4) for a particle immersed into an infinite medium, then $\mathbf{u}_1$ is determined by a boundary condition $\mathbf{u}_1|_S=-\mathbf{u}_0|_S$, the velocity $\mathbf{u}_2$ is determined by a boundary condition $\mathbf{u}_2|_p=-\mathbf{u}_1|_p$, and so on.

In the first nonvanishing order with respect to the parameter a/Δ, the expression for fluid velocity $\mathbf{u}_0$ reads (see Ref. 9, Chap. 1, Sec. 15)

$$\mathbf{u}_0=-A(\hat{s}\cdot\mathbf{k}), \tag{10}$$

where the tensor $\hat{s}$ is as follows:

$$\hat{s}=\frac{1}{|\mathbf{r}-\mathbf{c}|}\left(\hat{e}+\mathbf{nn}+(\hat{e}-3\mathbf{nn})\frac{a^2}{|\mathbf{r}-\mathbf{c}|^2}\right),$$

$$\mathbf{n}=\frac{\mathbf{r}-\mathbf{c}}{|\mathbf{r}-\mathbf{c}|}.$$

Coefficient A is determined from the last expression in the boundary conditions (4):

$$A=\frac{a\vartheta\xi_\kappa}{2}\frac{\rho_0}{(2\Delta)^2}.$$

The force $\mathbf{F}$ acting at the particle is determined by the following expression (see Ref. 9, Chap. 1, Sec. 15):

$$\mathbf{F}=8\pi\eta A\mathbf{k}. \tag{11}$$

In particular cases when heat sources are distributed uniformly over the surface, $q=q_S\delta(r-a)$, or over the volume, $q=q_V$ of the particle, $\delta(x)$ is Dirac's delta function

$$\mathbf{F}=\mathbf{k}\frac{\pi a\eta\vartheta}{\kappa_1\Delta^2}\frac{\kappa_1-\kappa_2}{\kappa_1+\kappa_2}Q, \tag{12}$$

where $Q=a^2q_S$ for a surface heat source, and $Q=(a^3/3)q_V$ for a volumetric heat source.

Equation (12) accounts for the temperature gradient induced at the particle embedded into the host medium when thermal conductivities of a host medium and an adjacent medium are different. If $\kappa_1=\kappa_2$, the system is homogeneous, and spatial temperature distribution is determined by spatial distribution of heat sources. In this study we consider an isotropic distribution of internal heat sources, and $\mathbf{F}=0$.

When $\kappa_1>\kappa_2$, Eqs. (5) and (7) imply the existence of the additional heat source with the same sign as a real heat source. Thus, a heat releasing particle is repelled from the surface, and heat absorbing particle is attracted to the surface. Indeed, the last in Eqs. (4) shows that the velocity of the fluid has the same sign as an induced temperature gradient. Since the direction of the particle velocity is the same as that of the surrounding fluid, the particle will move in the direction of the induced temperature gradient.

Similarly, when $\kappa_1<\kappa_2$, Eqs. (5) and (7) imply the existence of the additional heat source with the sign opposite to the sign of a real heat source. Thus, e.g., in the case of a heat releasing particle there occurs the induced heat absorption source located at $\mathcal{R}\mathbf{c}$. Therefore the induced temperature gradient, $\boldsymbol{\nabla}T\propto-\mathbf{k}$, and heat releasing particle is attracted to the surface. The situation is reversed in the case of a heat absorbing particle.

Equation (12) implies that a heat releasing particle is attracted to the interface when thermal conductivity of the host medium is less than thermal conductivity of the adjacent medium. Thus, e.g., growing in air by the condensation particle is attracted to a metal surface while an evaporating in air particle is repelled from a metal surface.

Certainly the result expressed by Eq. (12) remains only qualitative without determining a dependence of the coefficient ϑ upon the parameters of the problem. Kinetic theory (see Ref. 4) yields the following formula for this coefficient:

$$\vartheta=\frac{3}{4}\frac{\nu}{T_S}, \tag{13}$$

where $\nu=\eta/\rho$ is a kinematic viscosity.

When a particle itself is a heat source, a temperature at the particle surface T_S depends upon the power of a heat source, and upon the thermal conductivity of the particle, κ_0. Temperature distribution in a stationary regime is determined by the first equation in Eq. (1) where boundary conditions (3) at the interface must be supplemented with boundary conditions at the surface of the particle. Consider a case when a heat source has a spherical symmetry, i.e., $q(r)=q_V\theta(a-|\mathbf{r}-\mathbf{c}|)+q_S\delta(a-|\mathbf{r}-\mathbf{c}|)$, where q_V is the density of a uniform volumetric heat source, q_S is the density of a surface heat source, and $\theta(x)$ is the Heaviside step function. Then boundary conditions at the surface of the particle read

$$[T]_p=0,\quad \mathbf{n}\cdot[\kappa\boldsymbol{\nabla}T]_p=-q_S. \tag{14}$$

Solving this boundary value problem by the method of reflections as described above we arrive at the following expression for temperature T:

$$T = T^0 + T' + T''. \tag{15}$$

The term T^0 in Eq. (15) is determined by Eq. (1) and boundary condition (14). The term T' in Eq. (15) is determined by Eq. (1) with $q=0$ and with the following boundary conditions:

$$\begin{aligned} &\mathbf{n}\cdot[\kappa\boldsymbol{\nabla}T']_S = (\kappa_2-\kappa_1)(\mathbf{n}\cdot\nabla T^0)_S, \\ &[T']_S = 0, \;\; T'_\infty = 0. \end{aligned} \tag{16}$$

The term T'' in Eq. (15) is determined by Eq. (1) with $q=0$ and with the following boundary conditions:

$$\begin{aligned} &\mathbf{n}\cdot[\kappa\boldsymbol{\nabla}T'']_p = (\kappa_0-\kappa_1)(\mathbf{n}\cdot\nabla T')_p, \\ &[T'']_p = 0, \quad T''_\infty = 0. \end{aligned} \tag{17}$$

Denote by subscript 0 temperature inside a particle and by subscript 1 temperature of a host fluid. Then

$$\begin{aligned} &T_0^0 = \frac{q_V a^2}{3}\left[\frac{1}{\kappa_1} + \frac{1}{2\kappa_0}\left(1-\frac{r^2}{a^2}\right)\right] + \frac{q_S a}{\kappa_1} + \overline{T}, \\ &r = |\mathbf{r}-\mathbf{c}|, \\ &T_1^0 = \frac{a^2}{\kappa_1}\frac{\overline{Q}}{|\mathbf{r}-\mathbf{c}|} + \overline{T}, \quad \overline{Q} = q_S + \frac{1}{3}aq_V, \end{aligned} \tag{18}$$

where $\overline{T}$ is the initial temperature before introducing a heat source. The temperature at the particle's surface

$$T_S = \frac{\overline{Q}a}{\kappa_1} + \overline{T} \tag{19}$$

and

$$\begin{aligned} &T' = \frac{\xi_\kappa \overline{Q} a^2}{\kappa_1}\frac{1}{|\mathbf{r}-\hat{R}\mathbf{c}|}, \\ &T''_0 = \frac{\kappa_1-\kappa_0}{\kappa_0+2\kappa_1}(\boldsymbol{\beta}\cdot(\mathbf{r}-\mathbf{c})), \\ &T''_1 = \frac{\kappa_1-\kappa_0}{\kappa_0+2\kappa_1}\frac{a^3}{|\mathbf{r}-\mathbf{c}|^3}(\boldsymbol{\beta}\cdot(\mathbf{r}-\mathbf{c})), \end{aligned} \tag{20}$$

where

$$\beta = -\frac{\xi_\kappa Q a^2}{\kappa_1}\frac{\mathbf{k}}{(2\Delta)^2}, \quad \mathbf{k} = \frac{\mathbf{c}}{\Delta}.$$

Equations (4), (10), (11), (13), and (20) yield the following formula for the force $\mathbf{F}$:

$$\mathbf{F} = \frac{3\pi}{4}\frac{a^2}{\Delta^2}\frac{\nu^2\rho\xi_\kappa f}{\left(\dfrac{aQ_S}{\kappa_1}+\overline{T}\right)}\frac{\overline{Q}a}{\kappa_1}\mathbf{k}, \quad f = \frac{3\kappa_1}{\kappa_0+2\kappa_1}. \tag{21}$$

In the case of a "regular" thermophoresis associated with the external temperature gradient $(\boldsymbol{\nabla}T)_{\text{ext}}$ a thermophoretic force is determined by the following formula (see, e.g., Ref. 9, Chap. 1, Sec. 14):

$$\mathbf{F}_{\text{ext}} = -3\pi f\frac{a\nu^2\rho}{\overline{T}}(\boldsymbol{\nabla}T)_{\text{ext}}. \tag{22}$$

Let L be a characteristic length scale of variation of the external temperature. Then $(\boldsymbol{\nabla}T)_{\text{ext}} \sim \overline{T}/L$ and

$$\begin{aligned} &\frac{F}{F_{\text{ext}}} = \frac{1}{4}\xi_\kappa\frac{aL}{\Delta^2}\frac{a\overline{Q}}{\kappa_1\overline{T}}, \quad \text{if } \frac{a\overline{Q}}{\kappa_1\overline{T}} \ll 1, \\ &\frac{F}{F_{\text{ext}}} = \frac{1}{4}\xi_\kappa\frac{aL}{\Delta^2}, \quad \text{if } \frac{a\overline{Q}}{\kappa_1\overline{T}} \gg 1. \end{aligned} \tag{23}$$

Is must be noted that although in this study we considered boundary conditions for rigid particles [see Eq. (4)] the obtained results are valid (at least qualitatively) for small liquid droplets with large viscosity in comparison with that of a host medium. In view of the latter remark let us consider the magnitude of the investigated phenomenon using an example of an evaporating droplet of alcohol in air at temperature $\overline{T}=373$ K ($\overline{Q}=q_0\rho\dot{a}$, where q_0 is a latent heat of evaporation) $a\overline{Q}/\kappa_1\overline{T}\sim 0.1$. When external temperature gradient is small, i.e., $L \gg \Delta^2/a$, the self-induced thermophoretic force can be larger than a thermophoretic force caused by an external temperature gradient. Note that a thermophoretic force given by Eq. (18) $\mathbf{F}_{\text{ext}} \sim f(a/L)\nu^2\rho$, where $0 \leqslant f \leqslant 3/2$ and the ratio a/L can be varied in the experiments. The size of a particle when a force $\mathbf{F}\sim\nu^2\rho$ exceeds the gravitational force is of the order of $a \leqslant (\nu^2/g)^{1/3}(\rho/\rho_p)^{1/3}$, where g is the acceleration of gravity and ρ_p is a material density of the particle. Thus, e.g., in air under normal conditions this size is on the order of $a \leqslant 0.03$ cm.

Although the obtained results were derived under the assumption that $a \ll \Delta$, they are essentially valid also for $a/\Delta \sim 1$. It is known that a thermophoretic force increases indefinitely when a particle approaches a surface,[8] and the smaller is a Knudsen number $\text{Kn}=\lambda/a$, where λ is a free path length of gas molecules, the steeper the increase of the thermophoretic force. In the present study it is assumed that $\text{Kn} \ll 1$. The reason for the increase of a thermophoretic force is the increase of a temperature gradient when a particle approaches a surface. When a heat releasing particle approaches a surface, a temperature gradient also increases. Analysis of the behavior of a thermophoretic force acting on a heat releasing particle near the surface when $a/\Delta \sim 1$ requires numerical calculations and was not performed before.

III. THERMOPHORETIC INTERACTION OF HEAT RELEASING PARTICLES

One of the direct consequences of the above considered phenomenon of thermophoretic self action of a heat releasing particle is the existence of the effect of attraction in a system consisting of a small particle with the radius R_a and internal heat source q_a and a large particle with the radius R_b and internal heat source q_b (see Fig. 2). Denote thermal conductivities of the particles κ_a and κ_b, and assume that $\kappa_b \gg \kappa_1$, where κ_1 is thermal conductivity of a host medium. Temperature gradient induced by a particle a at the surface of a particle b, $\boldsymbol{\nabla}T_a^0(\{b\})$, results at the flow slip at the surface of the particle b. If the particle b is immobilized, then there is a force $\mathbf{F}_b^0$ acting at the particle due to the viscosity of a host medium. Similarly, a force $\mathbf{F}_a^0$ acts at the particle a due to an

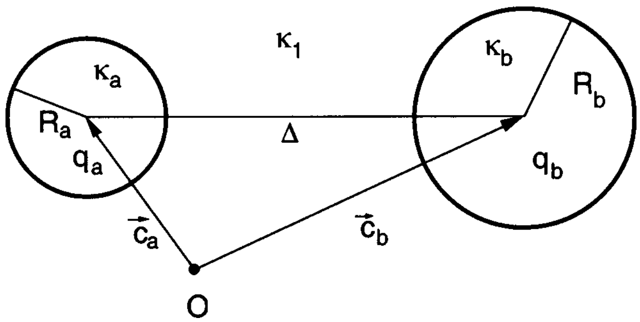

FIG. 2. Locations of particles a and b in a host medium with thermal conductivity κ_1. R_a, R_b and κ_a, κ_b are radii and thermal conductivities of particles a and b, respectively.

induced temperature gradient $\nabla T^0_b(\{a\})$. However, due to the thermophoretic self action which is caused by the induced temperatures $T^i_a(\{a\})$ and $T^i_b(\{b\})$, the particles will be attracted to each other if thermal conductivities of particles are larger than thermal conductivity of the host medium. Since the effect of the induced temperature is the effect of the higher order in the parameter R_a/Δ_{ab} and R_b/Δ_{ab}, where Δ_{ab} is a distance between the centers of the particles, the total effect is mutual repulsion of particles. However, if internal heat sources are such that $q_b \ll q_a$, then $\nabla T^i_a(\{a\}) \gg \nabla T^0_b(\{a\})$. In this case the self-induced attractive force acting at particle a is larger than a repulsive force produced by particle b. The resulting effect is that particle a is attracted to particle b. Let us determine these forces using a method of reflections.

Due to linearity of the problem temperature T as in the previous case can be represented as $T = T_a + T_b$, where

$$T_\sigma = T^0_\sigma + T'_\sigma + T''_\sigma. \tag{24}$$

Temperature T^0_σ, where $\sigma = a,b$ is determined by Eq. (1) and boundary conditions (14) at the surfaces of both particles. Thus temperature T^0_σ is determined by expression (18) where κ_0, $\overline{Q}$, a must be replaced by κ_σ, $\overline{Q}_\sigma$, R_σ.

Temperature T'_σ is determined by heat conduction equation with boundary conditions (16) where subscript S is replaced by subscript $\bar\sigma$. When $\sigma = a$ then $\bar\sigma = b$ and vice versa. For spherical particles with radii R_σ and $R_{\bar\sigma}$

$$T'_\sigma = D_\sigma(\kappa_1 - \kappa_{\bar\sigma}) \sum_{\ell=1}^{\infty} \frac{\ell}{\ell\kappa_{\bar\sigma} + (\ell+1)\kappa_1} \cdot \frac{R_{\bar\sigma}^{2\ell+1} \cdot P_\ell(\mu)}{\Delta^{\ell+1}|\mathbf{r} - \mathbf{c}_{\bar\sigma}|^{\ell+1}}, \tag{25}$$

where $\mu = \mathbf{k} \cdot \mathbf{n}_{\bar\sigma}$, $\mathbf{k} = (\mathbf{c}_\sigma - \mathbf{c}_{\bar\sigma})/\Delta$, $\Delta = |\mathbf{c}_\sigma - \mathbf{c}_{\bar\sigma}|$, $\mathbf{n}_\sigma = (\mathbf{r} - \mathbf{c}_{\bar\sigma})/|\mathbf{r} - \mathbf{c}_{\bar\sigma}|$, $D_\sigma = R^2_\sigma \overline{Q}_\sigma / \kappa_1$.

Expression (25) is a multipoles expansion of temperature T'_σ relative to a center of a particle $\bar\sigma$. This expression follows from Eq. (18) for T^0_1 and the known relation for a field of a point source near a sphere (for details see, e.g., Ref. 10, Problem 157). In order to determine expansion of temperature T'_σ into multipoles series with respect to a particle σ one can use Ref. 7 (Chap. 4, Sec. 7).

In the range $|\mathbf{r} - \mathbf{c}_\sigma| \ll \Delta$ in the leading order approximation with respect to the parameter $|\mathbf{r} - \mathbf{c}_\sigma|/\Delta \ll 1$ we find

$$\nabla T'_\sigma(\{\sigma\}) = -D_\sigma \frac{(\kappa_1 - \kappa_\sigma)^2}{\kappa_{\bar\sigma} + 2\kappa_1} \frac{R^3_{\bar\sigma}}{\Delta^5} \mathbf{k}. \tag{26}$$

In expression (26) we took into account that $R_\sigma \ll R_{\bar\sigma}$. Hereafter we assume that $R_a \ll R_b$.

A gradient of the temperature induced by a heat source b in the vicinity of a particle a can be determined using expression (18), and in the leading order approximation

$$\nabla T^0_b(\{a\}) = -\frac{R^2_b}{\kappa_1} \frac{\overline{Q}_b \mathbf{k}}{\Delta^2} = -\frac{D_b}{\Delta^2} \mathbf{k}.$$

In order to determine temperature T''_a we must use an equation similar to Eq. (17) when we have to take into account temperature gradient $\nabla T^0_b(\{a\})$, Thus for T''_σ we have the following boundary conditions:

$$[T'']_\sigma = 0, \quad [\kappa_\sigma(\mathbf{n} \cdot \nabla T'')]_\sigma = (\kappa_\sigma - \kappa_1)(\mathbf{n} \cdot \nabla)(T''_\sigma + T^0_{\bar\sigma}).$$

Denote

$$\tilde{\boldsymbol{\beta}}_\sigma = -\left(D_{\bar\sigma} + D_\sigma \frac{\kappa_1 - \kappa_{\tilde\sigma}}{\kappa_{\bar\sigma} + 2\kappa_1} \frac{2R^3_{\bar\sigma}}{\Delta^3} \right) \frac{\mathbf{k}}{\Delta^2}. \tag{27}$$

Then we obtain

$$T''_{0\sigma} = \frac{\kappa_1 - \kappa_\sigma}{\kappa_\sigma + 2\kappa_1} (\tilde{\boldsymbol{\beta}}_\sigma \cdot (\mathbf{r} - \mathbf{c}_\sigma)),$$

$$T''_{1\sigma} = \frac{\kappa_1 - \kappa_\sigma}{\kappa_\sigma + 2\kappa_1} \frac{R^3_\sigma}{|\mathbf{r} - \mathbf{c}_\sigma|^3} (\tilde{\boldsymbol{\beta}}_\sigma \cdot (\mathbf{r} - \mathbf{c}_\sigma)).$$

In this approximation the tangential component of the temperature gradient $(\boldsymbol{\tau} \cdot \nabla T)_\sigma$ is determined by the following expression:

$$(\boldsymbol{\tau} \cdot \nabla T)_\sigma = (\boldsymbol{\tau} \cdot \tilde{\boldsymbol{\beta}}_\sigma) f_\sigma, \quad f_\sigma = \frac{3\kappa_1}{\kappa_\sigma + 2\kappa_1}. \tag{28}$$

Inspection of the expression for $\tilde{\boldsymbol{\beta}}_\sigma$ Eq. (27) yields the following condition when temperature gradient at the particle's surface changes sign:

$$\frac{D_{\bar\sigma}}{D_\sigma} < \frac{\kappa_{\bar\sigma} - \kappa_1}{\kappa_{\bar\sigma} + 2\kappa_1} \frac{R^3_{\bar\sigma}}{\Delta^3}$$

or

$$\overline{Q}_{\bar\sigma} < \frac{R^2_\sigma R_{\bar\sigma}}{\Delta^3} \overline{Q}_\sigma \xi_\sigma, \quad \xi_\sigma = \frac{\kappa_{\bar\sigma} - \kappa_1}{\kappa_{\bar\sigma} + 2\kappa_1}. \tag{29}$$

It is possible to continue the iteration procedure keeping only the leading order term at each iteration. Such a procedure can be termed a procedure of leading order iterations. Since every particle is characterized by its parameters, the effect induced by one particle at the current iteration may exceed the effect induced by another particle at the preceding iteration. It is exactly a condition Eq. (29) that describes a situation when at the first iteration the effect of particle σ prevails over the effect of particle $\bar\sigma$ at the zeroth iteration.

Let us determine the forces exerted by fluid at the particles when particles are in rest in the laboratory frame of reference. As it was noted above this implies that there is a force $\mathbf{F}$ which is applied at the particle and keeps it at rest. Thermophoretic force that is calculated in this study, $\mathbf{F}_T = -\mathbf{F}$. Similarly to Sec. II for simplicity we consider the problem in the zeroth order approximation in Knudsen number Kn. The velocity field is determined by Eq. (1) with the following boundary conditions:

$$(\boldsymbol{\tau}\cdot\mathbf{u})_\sigma = \theta_\sigma(\boldsymbol{\tau}\cdot\nabla T)_\sigma, \quad (\mathbf{n}\cdot\mathbf{u})_\sigma = 0, \quad \mathbf{u}_\infty = 0. \tag{30}$$

Here θ_σ is determined by formula (13) where T_S is assumed to be equal to the temperature of the host medium without particles. The latter assumption is valid when the heat release rates are not high, so that according to Eq. (19) $T_S = \overline{T}$.

Fluid velocity is given by superposition $\mathbf{u} = \mathbf{u}_a + \mathbf{u}_b$, where $\mathbf{u}_\sigma = \mathbf{u}_\sigma^0 + \mathbf{u}_\sigma' + \mathbf{u}_\sigma''$. Each iteration $\mathbf{u}^n$ is determined by the equations $\eta\nabla^2(\nabla\times\mathbf{u}^n) = 0$, $\nabla\cdot\mathbf{u}^n = 0$ with corresponding boundary conditions. Thus, the term $\mathbf{u}_\sigma^0$ satisfies boundary conditions (30) at the surface of particle σ and

$$\mathbf{u}_\sigma^n(\{\sigma\}) = -\mathbf{u}_{\bar\sigma}^{n-1}(\{\sigma\}), \tag{31}$$

where $n > 0$ and $\{\sigma\}$ denotes coordinates at the surface of particle σ. Since the particles are kept at rest the only characteristic direction in the problem is determined by a vector $\mathbf{k}$. Expression for a velocity $\mathbf{u}_\sigma^n$ can be written as follows (see Ref. 9, Chap. 1, Sec. 15)

$$\mathbf{u}_\sigma^n = \frac{1}{|\mathbf{r}-\mathbf{c}_\sigma|}\left(a_\sigma^n \hat{S}_1 + \frac{b_\sigma^n R_\sigma^2}{|\mathbf{r}-\mathbf{c}_\sigma|^2}\hat{S}_2\right)\cdot\mathbf{k}, \tag{32}$$

where $\hat{S}_1 = \hat{e} + \mathbf{n}_\sigma\mathbf{n}_\sigma$, $\hat{S}_2 = \hat{e} - 3\mathbf{n}_\sigma\mathbf{n}_\sigma$. Since the problem is a linear one the total force acting at the particle can be written as $F_\sigma = \Sigma F_\sigma^n$, and $F_\sigma^n = -8\pi\eta a_\sigma^n$ (see Ref. 5, Chap. 2, Sec. 20). In order to transform the multipoles expansion with respect to a particle σ into a multipoles expansion with respect to a particle $\bar\sigma$, we will use the following formulas:

$$\mathbf{n}_{\bar\sigma} = (\mathbf{n}_\sigma t' + \mathbf{k})\sum_{\ell=0}^{\infty} P_\ell(\mu')(-1)^\ell t'^\ell, \quad t' = |\mathbf{r}-\mathbf{c}_\sigma|/\Delta,$$

$$\mathbf{n}_\sigma = (\mathbf{n}_{\bar\sigma} t - \mathbf{k})\sum_{\ell=0}^{\infty} P_\ell(\mu)(-1)^\ell t^\ell, \quad t = |\mathbf{r}-\mathbf{c}_{\bar\sigma}|/\Delta,$$

where $\mu' = \mathbf{k}\cdot\mathbf{n}_\sigma$ and $\mu = \mathbf{k}\cdot\mathbf{n}_{\bar\sigma}$.

In the first nonvanishing approximation with respect to parameters t' and t we arrive at the following expression for the velocity:

$$\mathbf{u}_{\bar\sigma}^{n-1}(\{\sigma\}) = 2\frac{a_{\bar\sigma}^{n-1}}{\Delta}\mathbf{k}.$$

Then boundary condition (30) and formula (31) yield

$$a_\sigma^n = -\frac{3}{2}\frac{R_\sigma}{\Delta}a_{\bar\sigma}^{n-1}, \quad b_\sigma^n = -\frac{1}{2}\frac{R_\sigma}{\Delta}a_{\bar\sigma}^{n-1}.$$

Thus in the leading order approximation the following force acts at particle a:

$$\mathbf{F}_a = \mathbf{F}_a^0 - \frac{3}{2}\frac{R_a}{\Delta}\mathbf{F}_b^0. \tag{33}$$

Here the force $\mathbf{F}_a^0$ is caused by the temperature gradient at the surface of particle a and it is given by the following expression: $\mathbf{F}_a^0 = -(3\pi\nu^2\rho/\overline{T})\boldsymbol{\beta}_a R_a f_a$

or

$$\mathbf{F}_a^0 = \frac{3\pi\nu^2\rho}{\overline{T}} f_a \frac{R_a}{\Delta^2}\left(\frac{R_b^2\overline{Q}_b}{\kappa_1} + \frac{R_a^2\overline{Q}_a}{\kappa_1}\frac{k_1-\kappa_b}{\kappa_b+2\kappa_1}2\frac{R_b^3}{\Delta^3}\right)\mathbf{k},$$
$$f_a = \frac{3\kappa_1}{\kappa_a+2\kappa_1}. \tag{34}$$

The force given by expression (34) comprises two terms. The first term describes a force induced by an external source b while the second term describes a self-induced thermophoretic force. The second term in expression (33),

$$\mathbf{F}_a' = -\frac{3}{2}\frac{R_a}{\Delta}\mathbf{F}_b^0,$$

is caused by a fluid flow induced by a temperature gradient at particle b. If condition (29) (where subscript $\bar\sigma$ corresponds to a particle b) is satisfied, then contribution of a self-induced temperature gradient into a force $\mathbf{F}_b^0$ can be neglected ($\overline{Q}_b \ll \overline{Q}_a$) and

$$\mathbf{F}_b^0 = \frac{3\pi\nu^2\rho}{\overline{T}} f_b R_b \frac{R_a^2\overline{Q}_a}{\kappa_1\Delta^2}\mathbf{k}. \tag{35}$$

Expression (35) determines a force acting at a particle b due to the temperature gradient at its surface induced by a particle a. Expressions (33)–(35) provide a solution of the problem. We do not present here a general formula that can be obtained by substituting Eqs. (34)–(35) into Eq. (33), and consider conditions when a force acting at a particle a is attractive. These conditions comprise a condition (29) that can be rewritten as

$$\overline{Q}_b < 2\overline{Q}_a\frac{\kappa_b-\kappa_1}{\kappa_b+2\kappa_1}\frac{R_a^2R_b}{\Delta^3} \tag{36}$$

and a condition

$$|\mathbf{F}_a^0| > \frac{3}{2}\frac{R_a}{\Delta}|\mathbf{F}_b^0|$$

[see Eq. (33)]. If condition (36) is satisfied within a wide margin of safety, then the first term in the right-hand side of Eq. (34) can be neglected (e.g., $\overline{Q}_b \cong 0$) and the condition for attraction reads

$$\frac{f_a}{f_b}\xi_b\frac{R_b^2}{\Delta^2} > \frac{3}{2}. \tag{37}$$

The latter condition corresponds to a case when a self-induced by a particle a thermophoretic force (attracting force) is larger than a thermophoretic force acting at particle a due to a fluid flow caused by a temperature gradient at a particle b (repelling force). Thus, a heat releasing particle with a low thermal conductivity is attracted to the particle with a high thermal conductivity. In the limiting case when $\kappa_b \gg \kappa_1$ and $\kappa_a \gg \kappa_1$, the condition for attraction Eq. (37) can be rewritten as

$$\frac{R_b^2}{\Delta^2} > \frac{3}{4}\frac{\kappa_a}{\kappa_b}. \tag{38}$$

It is interesting to note that in the range of the parameters indicated above neither condition (37) nor condition (38) depend upon the size of a particle a and a heat release rate. Since R_b is fixed, condition (37) determines the distance between the particles Δ where repulsion changes to attraction. In contrast to a case of a particle interacting with a plane surface where attraction occurs at any distance, attraction between spherical particles occurs only within a certain finite distance. The reason for this behavior is a fluid flow slip at the particle b which is caused by a heat source $\overline{Q}_a$. The higher the thermal conductivity of a particle b, κ_b, the lower the temperature gradient at the surface of a particle b that is induced by a heat source q_a and the smaller the repulsion force

$$\mathbf{F}_a' = -\frac{3}{2}\frac{R_a}{\Delta}\mathbf{F}_b^0.$$

Let us compare the magnitude of a thermophoretic force F_a^{atr} which is responsible for attraction of particle a [the second term formula (34)] with a classical thermophoretic force F_{ext} [Eq. (22)]. Since in Sec. II we have already compared a thermophoretic force F acting at a particle near the interface [Eq. (21)] with F_{ext} [see Eq. (23)], it is sufficient to compare a force F_a^{atr} with F. Equations (21) and (34) yield

$$\frac{F_a^{\text{atr}}}{F} = \frac{8R_b^3}{\Delta^3}\frac{\kappa_a+\kappa_1}{\kappa_b+2\kappa_1}. \tag{39}$$

In deriving Eq. (39) for consistency of notations we introduced the following changes in Eq. (21): $\kappa_2 \to \kappa_b$, $a \to R_a$, $\overline{Q} \to \overline{Q}_a$. In the most interesting case when $\kappa_b \gg \kappa_1$, $F_a^{\text{atr}} = (8R_b^3/\Delta^3)F$.

IV. CONCLUSIONS

We considered a thermophoretic force caused by the self action of a heat releasing or absorbing particles near the interface between two media with different thermal conductivities. This force is caused by the induced temperature gradient, which is proportional to the rate of heat release (absorption) by the particle. Therefore the magnitude of the thermophoretic force is proportional to the rate of heat release (absorption) by the particle, and its direction depends upon the sign of the parameter $\kappa_1 - \kappa_2$, where κ_1 is thermal conductivity of a host medium and κ_2 is thermal conductivity of the adjacent medium. The obtained results imply that a heat releasing particle is attracted to the interface when thermal conductivity of a host medium is less than thermal conductivity of the adjacent medium. Thus, e.g., growing in air by condensation particle is attracted to a metal surface while an evaporating in air particle is repelled from a metal surface.

We determined a condition for mutual attraction of heat releasing particles and derived an expression for the thermophoretic force acting at the particle. In contrast to a case of a particle interacting with a plane surface where attraction occurs at any distance, attraction between spherical particles occurs only within a certain finite distance.

It must be noted that thermophoretic interaction of particles was considered in earlier studies (see, e.g., Refs. 11–14 and references therein). However, a case with heat releasing particles was not considered before although it may play an important role in various phenomena, e.g., capture of condensing particles by a particle with a high thermal conductivity, capture of oxidizing particle by a larger particle (corrosion), etc.

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[1] J. Happel and H. Brenner, *Low Reynolds Number Hydrodynamics* (Martunus Nihhoff, Dordrecht, 1973).
[2] S. Kim and J. Karrila, *Microhydrodynamics: Principles and Selected Applications* (Butterworth-Heinemann, Boston, 1991).
[3] S. K. Friedlander, *Smoke, Dust, and Haze. Fundamentals of Aerosol Dynamics* (Oxford University Press, York, 2000).
[4] S. P. Bakanov, B. V. Deryagin, and V. I. Roldugin, Sov. Phys. Usp. **22**, 813 (1979) [Usp. Fiz. Nauk **129**, 255 (1979)]; S. P. Bakanov, Sov. Phys. Usp. **35**, 783 (1992) [Usp. Fiz. Nauk **162**, 133 (1992)].
[5] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Butterworth-Heinemann, Oxford, 1999).
[6] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984).
[7] E. W. Hobson, *The Theory of Spherical and Ellipsoidal Harmonics* (Cambridge University Press, Cambridge, 1965).
[8] Tatsuo Kanki, Heat Transfer-Jpn. Res. **27**, 57 (1998).
[9] E. M. Lifshitz and L. P. Pitaevsky, *Physical Kinetics* (Pergamon, Oxford, 1981).
[10] V. V. Batygin and I. N. Toptygin, *Problems in Electrodynamics* (Academic, London, 1978).
[11] H. J. Keh and S. H. Chen, Chem. Eng. Sci. **50**, 3395 (1995).
[12] S. H. Chen and H. J. Keh, J. Aerosol Sci. **26**, 429 (1995).
[13] Yu. Dolinsky and T. Elperin, Phys. Rev. E **64**, 061402(1–7) (2001).
[14] S. H. Chen, J. Aerosol Sci. **33**, 1155 (2002).