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DEPOSITION OF SMALL AEROSOL PARTICLES ON THE SURFACE OF MOVING
EVAPORATING CRYSTALS
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The method of augmenting asymptotic expansions is used for the case of low diffusive Peclet numbers to determine the flow of aerosol particles to the surface of an evaporating (or growing) crystal.

1. Formulation of the Problem. The theory of capture of small (moving in the freemolecular regime) aerosol particles by evaporating or condensing drops has by now been developed in quite an amount of detail [1-3]. As concerns processes of the capture of aerosols particles by evaporating or growing crystals, the theory is considerably less well developed.

A characteristic feature of particles of the solid phase (collectors) is their nonspherical form, which is considered in the present study.

We will examine a large (Knudsen number $K n=0$ ) evaporating particle of a solid phase suspended in a vapor-gas mixture. The theoretical analysis will be made for the case when the Reynolds number and the diffusive and thermal Peclet numbers are small, so that the equations of hydrodynamics and heat and mass transfer near the particle surface have the form

$$
\begin{equation*}
v \Delta \mathbf{v}=-\nabla p / \rho_{e}, \operatorname{div} \mathbf{v}=0, \Delta T_{e, i}=0, \Delta c_{1}=0 \tag{1.1}
\end{equation*}
$$

where $v, \rho_{e}, p$, and $T_{e}$ are the velocity, density, pressure, and temperature of the mixture; $c_{1}=n_{1} / n_{0} ; n_{0}=n_{1}+n_{2}$ ( $n_{1}$ and $n_{2}$ are the concentrations of the vapor and gas); $v$ is the kinematic viscosity of the mixture; $\mathrm{T}_{\mathrm{i}}$ is the particle temperature.

System (1.1) must be solved with allowance for the following conditions on the boundary between the particle (collector) and the medium:

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$$
\begin{gather*}
T_{e}=T_{i} \equiv T_{s}  \tag{1.2}\\
c_{1}=c_{1 s}\left(T_{s}\right)  \tag{1.3}\\
-\chi_{e} \partial T_{e} / \partial n=-\chi_{i} \partial T_{i} / \partial n-\rho_{e} v_{n} L  \tag{1.4}\\
n_{2} v_{n}+\frac{n_{0}^{2} m_{1}}{\rho_{e}} D_{12} \frac{\partial c_{1}}{\partial^{n}}=0  \tag{1.5}\\
v_{\tau}=0 \tag{1.6}
\end{gather*}
$$

and far from the particle:

$$
\begin{equation*}
\mathbf{v}=\mathbf{u}_{\infty}, c_{1}=c_{1 \infty} ; T_{e}=T_{\infty} \tag{1.7}
\end{equation*}
$$

In Eqs. (1.2)-(1.6), $D_{12}$ is the coefficient of binary diffusion; L, heat of the phase transition; $m_{1}$, mass of a vapor molecule; $x_{e}$ and $x_{i}$, thermal conductivities of the mixture and particle; $v_{n}$ and $v_{\tau}$, normal and tangential velocity components; $\partial / \partial n$, differentiation with respect to a normal to the particle (crystal) surface.

Equation (1.2) expresses the equality of the temperatures at the particle surface; Eq. (1.3) reflects the fact that the saturation vapor pressure at the particle surface is a function of temperature; Eq. (1.4) is derived from the law of conservation of energy; Eq. (1.5) expresses the impermeability of the surface of the evaporating particle to the gas molecules; Eq. (1.6) reflects the absence of gas slip along the particle surface.

The transport of small aerosol particles to the crystal (collector) surface is described by the convective diffusion equation

$$
\begin{equation*}
D \Delta N=\operatorname{u}_{\nabla} N \tag{1.8}
\end{equation*}
$$

where $D$ is the coefficient of Browian diffusion; $N$ is the concentration of particles; $\mathbf{u}$ is the velocity of the particles, which at low Reynolds numbers is due to motion of the collector and to Stefan flow caused by evaporation from the surface. It can be represented as

$$
\begin{equation*}
\mathbf{u}=\mathbf{v}+\mathbf{v}_{\mathbf{T}}+\mathbf{v}_{D}+\mathbf{v}_{\mathrm{st}} \tag{1.9}
\end{equation*}
$$

where $v$ is the gasdynamic velocity obtained from the solution of the problem of flow about a nonevaporating crystal; $\mathbf{v}_{\mathrm{T}}=-\mathrm{f}_{\mathrm{T}} \nabla \mathrm{T}$ is the rate of thermal phoresis; $\mathbf{v}_{\mathrm{D}}=-\mathrm{f}_{\mathrm{D}} \nabla \mathrm{c}_{1}$ is the rate of diffusive phoresis; $\mathbf{v}_{\text {St }}=-\left(\mathrm{n}_{0}^{2} \mathrm{~m}_{1} / \rho_{\mathrm{e}^{2}} \mathrm{n}_{2} \mathrm{D}_{12} \nabla \mathrm{c}_{1}\right.$ is the rate of Stefan flow.

The expressions for the functions $f_{T}$ and $f_{D}$ are cumbersome and can be found in works on thermal and diffusive phoresis [4, 5]. Henceforth, to abbreviate notation we will use the new function $f_{C}$, given by the relation $f_{C} \nabla c_{1}=f_{D} \nabla c_{1}+f_{T} \nabla T+v_{S t}$. We can do this because there is a one-to-one correspondence between the temperature and concentration gradients during evaporation of a particle.

We will change over to dimensionless variables in Eq. (1.8)

$$
\xi=\left(N-N_{\infty}\right) /\left(N_{a}-N_{\infty}\right), \quad \mathbf{r}_{*}=\mathbf{r} / a, \quad \mathbf{v}_{*}=\mathbf{v} / u_{\infty},
$$

where $\mathrm{N}_{\alpha}$ is the particle concentration at the surface (it is usually assumed that $\mathrm{N}_{a}=0$ ); $\alpha$ is a characteristic dimension of the particle-collector. In this case Eq. (1.8) reduces to the form

$$
\Delta \xi+\frac{f_{c}}{D} \nabla c_{1} \frac{\partial \xi}{\partial n}=\operatorname{Pev}_{*} \nabla \xi
$$

$\mathrm{Pe}=u_{\infty} \alpha / \mathrm{D}$, differentiation is done with respect to the dimensionless variables, and the function $\xi$ satisfies the following boundary conditions: $\xi_{r \rightarrow \infty}=0 ; \xi=\left.1\right|_{r=r_{0}}$, where $r_{0}$ is the radius-vector of the particle surface.

Henceforth, we will assume that the Peclet number $P e \ll 1$, while $P e_{1}=f_{c} / D$ is small: $P e_{1} \simeq 1$.
2. Calculation of the Flow of Aerosol Particles to the Surface. We will suppose that there is an orthogonal system of coordinates $x_{i}, x_{j}, x_{k}$ in which the particle surface is described by the equation $x_{i}=x_{0}$; in the same coordinate system the vapor concentration $c_{1}$ depends on the variable $x_{i}$, so that the Laplace equation of system (1.1) reduces to the equation

$$
\frac{d}{d x_{i}}\left[\theta\left(x_{i}\right) \frac{d c_{1}}{d x_{i}}\right]=0
$$

where $\theta\left(x_{i}\right)$ is a function of the variable $x_{i}$ determined from the relation [6]

$$
\psi\left(x_{j}, x_{h}\right) / \theta\left(x_{i}\right)=h_{j} h_{k} / h_{i}
$$

in which $h_{i}, h_{j}$, and $h_{k}$ are Lamé coefficients.
It should be noted that similar assumptions were used to construct a theory of diffusive charges of aerosol particles in [6].

We will use the method of augmenting asymptotic expansions to determine the concentration of aerosol particles. This method was explained in [7] with reference to hydrodynamics problems and in $[8,9]$ in regard to problems of heat and mass transfer.

We introduce external and internal expansions of the solution:

$$
\begin{gathered}
\xi^{*}=\sum_{n} \alpha^{(n)} \xi^{(n)}, \quad \lim _{\mathrm{Pe} \rightarrow 0} \frac{\alpha^{(n+1)}}{\alpha^{(n)}}=0 \\
\xi_{*}=\sum_{n} \alpha_{n} \xi_{n}, \quad \lim _{\mathrm{Pe} \rightarrow 0} \frac{\alpha_{n+1}}{\alpha_{n}}=0
\end{gathered}
$$

The internal expansion $\xi_{*}$ is valid for the internal flow zone $r_{0}<r_{*}<O\left(\mathrm{Pe}^{-1}\right)$, where $\mathbf{r}_{0}$ is the radius-vector of the particle surface. The external expansion is valid for the remaining part of the flow.

To determine the terms of the external expansion we introduce the condensed variable $\rho=$ Pe $\mathbf{r}_{*}$ and the velocity $\boldsymbol{\nabla}(\rho)=\mathbf{v}_{*}\left(\mathbf{r}_{*}\right)$. Henceforth the symbol $\%$ with the radius-vector and velocity will be omitted.

Since according to the above assumptions $P e_{1} \simeq 1$ and $c_{1} \simeq \beta / r$ for the harmonic function $c_{1}\left(x_{i}\right)$ as $r \rightarrow \infty$ (where $r$ is the radius of a certain spherical coordinate system connected with the particle), then to within the terms $O\left(\mathrm{Pe}^{2}\right)$ the equation (1.9) takes the following form for $\xi *$

$$
\Delta_{\rho} \xi^{*}=\mathbf{V}_{\nabla} \xi^{*}
$$

i.e., it coincides with the external expansion of the heat- and mass-transfer equation obtained in $[8,9]$.

It was shown in $[8,9]$ that to within $O\left(r^{-2}\right)$ the velocity field away from a particle of arbitrary form is

$$
\mathbf{v}=\mathbf{i}-\frac{3}{4 r}\left[\mathbf{F}+\frac{1}{r^{2}} \mathbf{r}(\mathbf{F r})\right]
$$

where $\mathbf{i}$ is a unit vector determining the direction of the flow; $\mathbf{F}$ is a dimensionless vector equal to the ratio of the resistance of the given particle to the Stokes force associated with the resistance of a solid spherical particle of radius $\alpha$.

Let us proceed to the construction of the solution.
Zeroth Approximation. The zeroth approximation for the external expansion is obvious: $\xi^{(0)} \frac{\text { Zeroth Approximation. The zeroth approximation }}{\equiv 0 \text {, while for the internal expansion with } \alpha_{0}=1 \text { we obtain the equation }}$

$$
\begin{equation*}
\Delta \xi_{0}+\frac{f_{c}}{D} \nabla c_{1} \frac{\partial \xi_{0}}{\partial n}=0 \tag{2.1}
\end{equation*}
$$

the solution of which, with allowance for the assumptions made earlier regarding the form of the particle, has the form

$$
\xi_{0}=b_{0} \exp \left[-\frac{f_{c}}{D} c_{1}(x)\right]+\frac{I_{0}}{A f_{c}}
$$

where $A=\frac{1}{\theta\left(x_{i}\right)} \frac{d c_{1}}{d x_{i}}=$ const; $\mathrm{b}_{0}$ and $\mathrm{I}_{0}$ are constants determined from the boundary conditions; meanwhile, Io determines the particle flow to the surface of a stationary collector accurately to within the constant factor. Expressions for these constants are easily obtained:

$$
\begin{gather*}
b_{0}=\frac{1}{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)}  \tag{2.2}\\
I_{0}=\frac{A f_{c}}{1-\exp \left[-\frac{f_{\mathrm{c}}}{D}\left(c_{1 s}-c_{1 \infty}\right)\right]}
\end{gather*}
$$

As $r \rightarrow \infty$, the function $\xi_{0}$ has the form $\beta / r$. Thus, from the condition of augmentation of the expansions, $\alpha^{(1)}=P e$.

First Approximation. The external expansion of the solution $\xi^{(1)}$ satisfies the equation

$$
\Delta_{\rho} \xi \xi^{(1)}=i \nabla_{\rho} \xi^{(1)}
$$

the solution of which, meeting the augmentation conditions, has the form

$$
\xi^{(1)}=\frac{\gamma}{\rho} \exp \left[\frac{\rho}{2}(\mu-1)\right]
$$

where $\mu=$ (iF); $\gamma$ is a constant determined from the augmentation conditions as $\rho \rightarrow 0$.
With allowance for the constancy of the total particle flow $I$ to the surface and the augmentation conditions $\lim _{r \rightarrow \infty} \xi^{(0)}=\lim _{\rho \rightarrow 0} \xi^{(1)}$, it is not hard to find that $\gamma=-I_{0} / D$.

Expansion of the function $\xi(1)$ in the internal variables with $r \rightarrow 0$ leads to the following selection of the parameter of the expansion $\alpha^{(1)}$ and the augmentation condition:

$$
\begin{equation*}
\alpha^{(1)}=P e, \lim _{r \rightarrow \infty} \xi_{1}=-\frac{\gamma}{2}(\mu-1) \tag{2.3}
\end{equation*}
$$

The function $\xi_{1}$ satisfies the equation

$$
\begin{equation*}
\Delta \xi_{1}+\frac{f_{c}}{D} \frac{\partial c_{1}}{\partial n} \frac{\partial \xi_{1}}{\partial n}=\mathbf{v} \nabla \xi_{0} \tag{2.4}
\end{equation*}
$$

and the condition on the boundary $\xi_{1}\left(x_{0}\right)=0$.
The function $\xi_{1}$ can be represented in the form $\xi_{1}=\xi_{10}\left(x_{i}\right)+\xi_{11}\left(x_{i}, x_{j}\right.$, $\left.x_{k}\right)$, where the function $\xi_{I 0}\left(x_{i}\right)$, with allowance for conditions (2.3) and the condition on the boundary, has the form

$$
\begin{gather*}
\xi_{10}=b_{1} \exp \left[-\frac{f_{c}}{D} c_{1}\left(x_{i}\right)\right]+\frac{I_{1}}{A f_{c}}, \\
b_{1}=\frac{\gamma}{2\left[\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)\right]},  \tag{2.5}\\
\quad-\gamma A f_{c} \exp \left(-\frac{f_{c}}{D} c_{1 s}\right) \\
I_{1}=\frac{f_{c}}{2\left[\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)\right]},
\end{gather*}
$$

While the function $\xi_{11}$ satisfies nonhomogeneous equation (2.4) and, with allowance for the limitations on the geometry of the particles introduced above, does not contribute to the total particle flow to the collector surface.

Analysis of the asymptote of the expression for $\xi_{1}$ with $r \rightarrow \infty$ shows that the parameter of the expansion $\alpha^{(2)}=$ Pe.

The equation satisfied by the function $\xi^{(2)}$ has the form

$$
\left(\Delta_{\rho}-\frac{1}{\mu} \frac{\partial}{\partial \rho}-\frac{1}{1-\mu^{2}} \frac{\partial}{\partial \mu}\right) \xi^{(2)}=-\frac{3}{4} \gamma\left[\mathbf{F}+\frac{(\mathbf{F} \rho)}{\rho^{2}} \boldsymbol{\rho}\right] \nabla_{\rho}\left\{\frac{1}{\rho} \exp \left[-\frac{1}{2} \rho(1-\mu)\right]\right\}
$$

The asymptotic expression for the function $\xi^{(2)}$ with $\rho \rightarrow u$ has the form (2.6)

$$
\xi^{(2)} \rightarrow-(1 / 2) \gamma(\mathrm{Fi}) \ln \rho
$$

The presence of the logarithmic term in (2.6) changes the power nature of the functions $\alpha_{n}(\mathrm{Pe})$ to a logarithmic character and leads to the following definition of the parameter of the expansion $\alpha_{2}$ :

$$
\alpha_{2}=\mathrm{Pe}^{2} \ln \mathrm{Pe}
$$

Function $\xi_{2}$ again satisfies Eq. (2.1) with boundary conditions:

$$
\xi_{2}\left(x_{0}\right)=0, \quad \lim _{r \rightarrow \infty} \xi_{2}=-\frac{1}{2} \gamma(\mathbf{F i})
$$



Fig. 1


Fig. 2

Using Eqs. (2.2), (2.5), and (2.7), we can write relations which determine the dimensionless flow of particles to a moving collector:

$$
\begin{gather*}
\xi_{2}=\frac{(\mathbf{F i}) \gamma \exp \left[-\frac{f_{c}}{D} c_{1}(x)\right]}{2\left[\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{\mathbf{c}}}{D} c_{1 \infty}\right)\right]}-\frac{\gamma(\mathbf{F i}) A f_{c} \exp \left(-\frac{f_{c}}{D} c_{1 s}\right)}{2\left[\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)\right]} .  \tag{2.7}\\
\operatorname{Sh}=\operatorname{Sh}_{0}\left[1+\frac{\operatorname{Pe}}{2} \frac{A f_{c}}{D} \frac{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)}{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)}+\frac{\mathrm{Pe}^{2} \operatorname{lnPP}}{2}\left(\mathbf{F i} \frac{A f_{c}}{D} \frac{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)}{\left.\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)\right]}\right.\right.
\end{gather*}
$$

where $S_{0}$ is the Sherwood number for the particle flow to a stationary collector.
3. Example of Calculations and Analysis of Results. As an example we will present the expression for the Sherwood number calculated for an evaporating particle having the form of an oblate spheroid and moving normally with respect to the major semiaxis:

$$
\begin{gather*}
\mathrm{Sh}=\mathrm{Sh}_{0}\left[1+\frac{\mathrm{Pe}}{2} \frac{c_{1 \infty}-c_{1 s}}{\pi / 2-\operatorname{arctg} \sigma_{0}} \frac{f_{c}}{D} \frac{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)}{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)}+\frac{4}{3 \pi} \mathrm{Pe}^{2} \ln \mathrm{Pe}\right. \\
\left.\times \frac{c_{1 \infty}-c_{1 s}}{\pi / 2-\operatorname{arctg} \sigma_{0}} \frac{f_{c}}{D} \frac{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)}{\exp \left(-\frac{f_{c}}{D} c_{1 s}\right)-\exp \left(-\frac{f_{c}}{D} c_{1 \infty}\right)}\right]  \tag{3.1}\\
\mathrm{Sh}_{0}=\frac{c_{1 \infty}-c_{1 s}}{\pi / 2-\operatorname{arctg} \sigma_{0}} \frac{2 f_{c}}{D} \frac{\exp \left[\frac{f_{c}}{D}\left(c_{1 \infty}-c_{1 s}\right)\right]}{1-\exp \left[\frac{f_{c}}{D}\left(c_{1 \infty}-c_{1 s}\right)\right]} .
\end{gather*}
$$

Here, $\sigma$ is the "radial" coordinate of the spheroid, connected with Cartesian coordinates by the relations:

$$
\begin{gathered}
x=a \sqrt{1+\sigma^{2}} \sqrt{1-\tau^{2}} \cos \varphi, y=a=\sqrt{1+\sigma^{2}} \sqrt{1-\tau^{2}} \sin \varphi \\
z=a \sigma \tau
\end{gathered}
$$

where $0 \leqslant \varphi \leqslant 2 \pi,-1 \leqslant \tau \leqslant 1, \sigma \geqslant 0, \sigma=\sigma_{0}$ corresponds to the surface of the spheroid. Figure 1 shows results of calculation of the Sherwood number in relation to the relative moisture content performed by Eq. (3.1). The calculation was performed for ice crystals with an aspect ratio $\varepsilon=0.03$ falling in a gravitational field. Curves 1 and 2 correspond to a crystal with the major semiaxis size $\alpha=15$ and $10 \mu \mathrm{~m}$. The ambient temperature $\mathrm{T}_{\infty}=-10^{\circ} \mathrm{C}$, the radius of the small particles $R=10^{-2} \mu \mathrm{~m}$, and pressure in the vapor-gas mixture away from the crystal $p=9.8 \cdot 10^{4} \mathrm{~N} / \mathrm{m}^{2}$.

It is evident from Fig. 1 that the flow of particles to the surface of an evaporating crystal exceeds the magnitude of the particle flow to the surface of a crystal not undergoing a phase transition.

A similar conclusion was obtained experimentally in [10] for an evaporating drop. This result, at first unexpected, can be explained by the action of the thermophoretic force directed toward the crystal surface during its evaporation.

Figure 2 shows the dependence of the Sherwood number on the dimensions of the aerosol particles. The calculations were performed for an evaporating (relative moisture content $75 \%$ ) ice crystal with a major semiaxis $\varepsilon=9 \mu \mathrm{~m}$ falling in a gravitational field. Curves 1 and 2 correspond to the temperature $\mathrm{T}_{\infty}=-5$ and $-10^{\circ} \mathrm{C}$, while curve 3 corresponds to a relative moisture content of $100 \%\left(\mathrm{~T}_{\infty}=-5^{\circ} \mathrm{C}\right)$.

The results of the calculations show that the effect of thermal-diffusive electrophoretic forces caused by evaporation from the surface of a collector on the capture of aerosol particles depends significantly on the dimensions of the particles.

The effect of thermal-diffusive electrophoretic forces on the capture of aerosol particles with a radius $R<5 \cdot 10^{-3} \mu \mathrm{~m}$ can be ignored even in the case of a low relative moisture content (relative humidity).

At the same time, evaporation from the surface of a collector may significantly increase the efficiency of capture of aerosol particles with a radius $\mathrm{R}>10^{-2} \mu \mathrm{~m}$.

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