PHOTOPHORETIC CONTRIBUTION TO THE TRANSPORT OF ABSORBING PARTICLES ACROSS COMBUSTION GAS BOUNDARY LAYERS[1]

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Abstract

Since radiation energy fluxes can be comparable to 'convective' (Fourier) fluxes in large fossil-fuel-fired power stations and furnaces, we have examined particle drift ('phoresis') induced by nonuniform photon-particle heating in a 'host' gas. Our analysis (Mackowski, 1988) of the photophoretic velocity includes the important 'slip-flow' regime, and the numerical results show that photophoresis is a significant transport mechanism for micron-sized absorbing particles in high radiative transfer combustion environments, with equivalent photophoretic diffusivities (dimensionless photophoretic velocities) being as large as 10 percent of the better-known thermophoretic diffusivity (Rosner, 1980, 1985). Since pervious experimental results (Rosner and Kim, 1984) demonstrated that thermophoresis causes over a 3-decade increase in particle deposition rates by convective diffusion, clearly, for small, absorbing particles, photophoresis will also be an important contributor to observed deposition rates. Accordingly, we present mass transfer coefficients for particle transport across laminar gaseous boundary layers, including both particle thermophoresis and photophoresis.

Thermophoresis and Photophoresis

When both radiative and convective energy fluxes are present in a gas environment with a dilute amount of aerosol particles, the motion of these particles is affected by temperature gradients in two different ways.

Thermophoresis describes the phenomenon wherein small particles in a gas experience a force in the direction opposite to the thermal gradient in the gas. The thermophoretic velocity (*i.e.*, the terminal velocity reached for an isolated particle in a gas with a constant temperature gradient) is normally written in the form:

$$\vec{v}_{th} = \alpha_{th}\left[\frac{-(\text{grad } T)}{T}\right] \qquad [1]$$

where α_T is a dimensionless thermal diffusion factor and D_p the diffusion coefficient of the particles. Actually, D_p is included here just to emphasize the similarity between $\vec{v}_{th}$ and a diffusion

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velocity, but the value of the thermophoretic diffusivity $\alpha_T D_p$ does not really depend on Dp (which, in fact, will be taken to be zero in our analysis). Talbot *et al.* (1980), presented an expression for $\alpha_T D_p$:

$$\alpha_{th} D_p = \frac{2c_s \nu \left(\frac{K_g}{K_p} + c_t \frac{l}{a}\right)\left[1 + \frac{l}{a}\left(A + B\, e^{-Ga/l}\right)\right]}{\left(1 + 3\, c_m \frac{l}{a}\right)\left(1 + 2\frac{K_g}{K_p} + 2c_t \frac{l}{a}\right)} \qquad [2]$$

where K_g and K_p are the thermal conductivities of the gas and particle, respectively; l is the gas mean-free path, $l=2\nu/\bar{c}$, with $\bar{c}=(8RT/\pi)^{1/2}$; a is the particle radius, A=1.20, B=0.41, G=0.88, and, for perfect accommodation between the particle and the gas molecules, c_s=1.17, c_m=1.1 and c_t=2.18; ν is the gas kinematic viscosity.

Note that, in the limit of $Kp \geq Kg$ and $a << l$, Eq. [1.2] reduces to:

$$\alpha_{th} D_p \cong \frac{c_s \nu}{3c_m}(A + B) \cong 0.56\nu$$

(the numerical factor in Waldmann's theory, 1961, is 0.54). This simple limiting case provides a first approximation for the magnitude of $\alpha_{th} D_p$, and motivates our introduction of $\alpha \equiv \alpha_{th} D_p / \nu$ in the analysis and examples which follow.

On the other hand, photophoresis takes into account the particle motion induced by the temperature gradient upon the particle surface originating from the nonuniform absorption of the radiant energy within the particle. The correspondent photophoretic velocity for an isolated particle can be written as

$$\vec{v}_{ph} = \alpha_{ph}\, D_p \frac{\vec{q}_R}{K_g T} \qquad [3]$$

where $\alpha_{ph} D_{ph}$ is the photophoretic diffusivity and $\vec{q}_R$ the radiative heat flux. Mackowski (1988) obtained an expression for $\vec{v}_{ph}$ in the slip flow regime, resulting in

$$\alpha_{ph} D_p = -\frac{2c_s \nu \bar{J}_1}{3} \frac{1 + \frac{l}{a}\left(A + B\, e^{-Ga/l}\right)}{\left(1 + 3\, c_m \frac{l}{a}\right)\left(1 + 2c_t \frac{l}{a} + 2\frac{K_g}{K_p}\right)} \qquad [4]$$

whereas in the free molecular limit

$$\alpha_{ph} D_p = -0.14\, \nu\, \bar{J}_1 \frac{a}{l} \qquad [5]$$

$\bar{J}_1$ is the thermophoretic asymmetry factor and represents a weighted integration of the absorption of radiant energy over the particle volume. For spherical, homogeneous particles and monochromatic radiation, $\bar{J}_1$ can be obtained from Lorenz-Mie theory as a function of the particle radiative size parameter $x=2\pi a/\lambda$, where λ is the radiation wavelength, and the complex index of refraction $m=n+ik$. An exact, series-expansion expressions for $\bar{J}_1$ has been derived which is analogous to the expressions for the radiative cross sections (Mackowski, 1988). For spectrally-distributed radiation, $\bar{J}_1$ is obtained from integration over the wavelength distribution.

Realize that $\bar{J}_1$ can be positive or negative, leading to $\vec{v}_{ph}$ directed either against or with the incident radiation direction. For radiation absorbed entirely on the particle surface, $\bar{J}_1$ attains a minimum value of –0.5.

Underlying Assumptions

To simplify the problem without losing its essential features, the following defensible assumptions will be made:

A1. The flow within the BL is steady and laminar. The usual BL approximations will be used and self-similarity will be assumed (see, *e.g.*, Schlichting, 1968).

A2. The aerosol particles are very dilute so that the prevailing velocity and temperature field are not affected by their presence.

A3. All thermophysical properties of the gas will be considered constant and equal to the values for the carrier gas at mainstream conditions. Transport properties for the dispersed aerosol will also be taken to be constant. Lastly, the system will be considered effectively incompressible, *i.e.*, the density will be assumed to be constant.

A4. Aerosol particles do not appreciably Brownian diffuse. Therefore, at each position, the velocity of the particles is taken to be the gas velocity plus the thermophoretic and photophoretic velocities, with these velocities being those corresponding to an isolated particle in a uniform gas with the same temperature gradient and radiant energy flux. The direction of the radiative flux will be taken along the normal to the solid collecting surface.

We consider the two-dimensional stagnation point (Hiemenz) flow. This corresponds to a steady flow which arrives from the y-axis, impinges on a flat solid wall placed at y=0, where it divides into two streams near the wall, leaving in both (±) directions. The external (inviscid) velocity distribution in the neighborhood of the symmetrical forward stagnation "point" (at x=y=0) is given by (*e.g.* Schlichting, 1968):

$$u_e(x) = \left(\frac{du_e}{dx}\right)_{x=0} \cdot x \qquad [6]$$

In the immediate vicinity of the solid wall, viscous (momentum diffusion) effects become important and, for a Newtonian fluid, the velocity field must satisfy the well-known two-dimensional BL equations:

$$u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = u_e\frac{\partial u_e}{\partial x} + \nu\frac{\partial^2 u}{\partial y^2} \qquad [7]$$

and

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \qquad [8]$$

with boundary conditions: $u=0$ (no slip) and $v=0$ (no blowing) at $y=0$ and $u = u_e(x)$ at $y=\infty$.

Introducing the "stretched" dimensionless coordinate

$$\eta \equiv y\left[\frac{1}{v}\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2} \qquad [9]$$

and a stream function given by

$$\psi(x,y) = \left[v\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2} \cdot f(\eta) \cdot x \qquad [10]$$

the equation of local mass conservation [8] is automatically satisfied and the velocity components become

$$u = \frac{\partial\psi}{\partial y} = u_e(x) \cdot f'(\eta) \qquad [11]$$

and

$$v = -\frac{\partial\psi}{\partial x} = -\left[v\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2} \cdot f(\eta) \qquad [12]$$

where above, and in what follows, primes denote differentiation with respect to η. Introducing these expressions into the x-momentum balance, equation [7], the following well-known nonlinear third-order (Blasius) ODE for $f(\eta)$ is obtained:

$$f''' + ff'' + \left[1-(f')^2\right] = 0 \qquad [13]$$

with the boundary conditions:

$$f = f' = 0 \qquad @\ \eta = 0 \qquad [14]$$

$$f' = 1 \qquad @\ \eta = \infty \qquad [15]$$

Notice that our assumptions of constant thermophysical properties and low mass loading allow $f(\eta)$ to be determined independently of the temperature and mass-fraction fields discussed below. Indeed, we will make use of the previous numerical computations of this well-known (Blasius) function (Schlichting, 1968).

Temperature Field

In the steady state, using laminar BL approximations A1, the PDE which governs the temperature distribution $T(x,y)$ is

$$u\frac{\partial T}{\partial x} + v\frac{\partial T}{\partial y} = \alpha_h \frac{\partial^2 T}{\partial y^2} \qquad [16]$$

α_h being the heat (thermal) diffusivity. Defining

$$\theta \equiv \frac{T}{T_\infty} \qquad [17]$$

when the wall temperature, T_w, is held constant, the ODE for $\theta(\eta)$ becomes

$$\theta'' + Pr \cdot f(\eta) \theta' = 0 \qquad [18]$$

subject to the boundary conditions

$$\theta(0) \equiv \theta_w = \frac{T_w}{T_\infty}, \quad \theta(\infty) = 1 \qquad [19]$$

where Pr is the host gas Prandtl number, ν/α_h. The solution can be written in the following quadrature form (*e.g.* Spalding & Evans, 1961):

$$\theta(\eta) = \theta_w + (1-\theta_w) \cdot \frac{1}{\delta_T} \int_0^\eta \exp\left[-\Pr \int_0^\varphi f(\xi)\, d\xi\right] d\varphi \qquad [20]$$

where $f(\eta)$ is defined by [13]-[15] and

$$\delta_T \equiv \int_0^\infty \exp\left[-\Pr \int_0^\varphi f(\xi)\, d\xi\right] d\varphi \qquad [21]$$

For a description of the computation of δ_T see, for example, Castillo & Rosner (1988), Section 3.1. For Pr=0.7 (*e.g.* air) we find δ_T=2.01669.

Particle Number Density

Now consider that in the mainstream there are $N_{p,\infty}$ particles per unit volume, each with the same radius[5], a_∞. In the absence of particle coagulation or break-up, the number density of particles N_p satisfies the equation

$$\mathrm{div}(\vec{v}_p N_p) = 0 \qquad [22]$$

Under assumption A4., the local particle velocity is given by

$$\vec{v}_p = \vec{v} + \vec{v}_{ph} + \vec{v}_{th} \qquad [23]$$

with $\vec{v}_{ph}$ and $\vec{v}_{th}$ given by Eq.[3] and Eq.[1], respectively. Defining $n \equiv N_p/N_{p,\infty}$, the first ODE for n takes the simple form

$$A\frac{dn}{d\eta} + Bn = 0 \qquad [24]$$

where we have introduced the dimensionless functions

$$A(\eta) \equiv f(\eta) + \frac{1}{\theta}\left(\beta + \alpha\frac{d\theta}{d\eta}\right) \qquad [25]$$

$$B(\eta) \equiv \frac{d}{d\eta}\left\{\frac{1}{\theta}\left(\beta + \alpha\frac{d\theta}{d\eta}\right)\right\} \qquad [26]$$

with

$$\alpha \equiv \frac{\alpha_{th} D_p}{\nu} \qquad [27]$$

$$\beta \equiv -\frac{\alpha_{ph} D_p q_R}{K_g T_\infty}\left[\left(\frac{du_e}{dx}\right)_{x=0}\nu\right]^{-1/2} \qquad [28]$$

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[5] This assumption may easily be relaxed in order to deal with a distribution of particle sizes in the main stream.

The solution of Eq.[24], with boundary condition n=1 @ $\eta=\infty$, can be written in the form of a quadrature

$$n = \exp\left[\int_{\eta}^{\infty} \frac{B(\varphi)}{A(\varphi)} d\varphi\right] \qquad [29]$$

The normal velocity of particles within the boundary layer is given by

$$v_p = v + v_{th} + v_{ph} = -\left[\nu\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2}\left[f + \alpha\left(\frac{d\ln\theta}{d\eta}\right) + \frac{\beta}{\theta}\right] \qquad [30]$$

which is negative for particles approaching the wall.

Under some circumstances, the particles do not arrive to the wall and a dust free region appears inside the boundary layer (Goren, 1977). The separation line between the region with particles and the dust free zone is located at the value of η where $v_p=0$; that is

$$A = f + \frac{1}{\theta}\left(\beta + \alpha\frac{d\theta}{d\eta}\right) = 0$$

When thermophoresis and photophoresis both push the particles away from the wall, that is, when $\theta_w>1$ and $\beta<0$, the dust free zone will exist for any value of β and θ_w and particles will not be collected by the solid surface. On the other hand, when both transport mechanisms compete in bringing particles towards the wall, the dust free zone exists only when

$$-\beta \geq \frac{\alpha}{\delta}\left(1 - \theta_w\right) \quad \text{for } \beta < 0 \quad \text{and } \theta_w < 1$$

or

$$\frac{\alpha}{\delta}\left(\theta_w - 1\right) \geq \beta \qquad \text{for } \beta>0 \quad \text{and } \theta_w > 1$$

Note that when both transport velocities oppose each other and are exactly equal, in modulus, at the wall (i.e., when the equal sign is verified in the above inequalities), the separation line coincides with the wall; that is, the deposition of particles vanishes even when the particles are everywhere inside the boundary layer. In this very particular case, however, some deposition will occur due to Brownian diffusion.

Anyway, here we are mainly interested in the cases when none of the above inequalities holds and deposition of particles takes place. When the particles arrive to the wall, the deposition rate of particles is given by

$$-N_w\, v_{p,w} = \left[\nu\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2} N_\infty \frac{n_w}{\theta_w}\left[\beta + \frac{\alpha}{\delta}\left(1 - \theta_w\right)\right] \qquad [31]$$

Thus, the dimensionless capture fraction, S, of particles will be

$$S \equiv \frac{-N_w\, v_{p,w}}{N_\infty\left[\nu\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2}} = \frac{n_w}{\theta_w}\left[\beta + \frac{\alpha}{\delta}\left(1 - \theta_w\right)\right] \qquad [32]$$

When we are interested in mass deposition rate instead of particle deposition, the relevant parameter is

$$J_m = \frac{1}{\rho\omega_\infty}\left[\nu\left(\frac{du_e}{dx}\right)_{x=0}\right]^{1/2}\left(-j''_{m,w}\right) \qquad [33]$$

where ω_∞ is the mass fraction of particles at mainstream and $\left(-j''_{m,w}\right)$ is the mass deposition rate at the wall. It is easy to see that J_m=S. When we consider the more practical case of a distribution of particle sizes at mainstream, the definition [33] is still valid, now with ω_∞ and $j''_{m,w}$ taking into account the contribution of the different sizes. In that case, it results in

$$J_m = \frac{\int N_\infty(a)\, a^3\, S(a)\, da}{\int N_\infty(a)\, a^3\, da} \qquad [34]$$

with S(a) given by [32] and n_w, β and α being functions of the particle radius a.

Results for Simultaneous Photophoresis and Thermophoresis

Figure 1 represents the dimensionless capture fraction, S, as a function of the ratio T_w/T_∞, for particles having a thermophoretic coefficient α=0.5 (a value close to the free molecular limit). The line for β=0 corresponds to pure thermophoretic deposition with negligible photophoretic transport. In that case, particles are captured only by cold surfaces (*i.e.* when $T_w/T_\infty<1$) and the deposition rate increases as the wall temperature decreases. When photophoresis helps to bring the particles towards the surface (*i.e.* when $\beta>0$), it produces two effects: on one hand, it allows the capture of particles even for moderate hot surface ($T_w/T_\infty>1$) and on the other hand it considerably increases the value of S for a given value of the ratio T_w/T_∞. The opposite is true for negative values of β, the range of temperatures over which deposition occurs is diminished as well as the deposition rate for a given temperature. Analogous results are obtained for α=0.1 (Figure 2) although the relative importance of photophoresis is higher. Thus, for $\beta=-10^{-1}$, no deposition occurs for the entire range of temperatures considered.

From Eq.[28], it can be seen that

$$\beta = \frac{1-\theta_w}{\delta}\left(\frac{q_R}{q_F}\right)\alpha F$$

where q_F is the conductive (Fourier) heat flux at the solid surface, and F is the ratio v_{ph}/v_{th} computed for equal q_R and q_F. For particles in the slip-flow regime (l/a<1), F can be expressed by

$$F = \frac{\bar{J}_1 K_g}{3 K_p\left(\frac{K_g}{K_p} + c_t \frac{l}{a}\right)}$$

Numerical results of F for carbonaceous char and fly-ash particles exposed to a black body radiation spectrum at T_{Rad}=1800K have been presented by Mackowski (1988) and are reproduced in Fig.3.

By using the above expression for β together with the values of F indicated in Fig.3, the deposition rate of char particles has been obtained and it is represented in Figure 4 for a fixed value of T_w/T_∞=0.7 and different ratios of q_R/q_F. For vanishing radiative fluxes the larger char particles are more efficiently captured due to their larger thermophoretic coefficient α (obtained from Eq.[2]). When the radiative heat flux is directed from the solid surface towards the bulk (*i.e.*, when $q_R<0$), the char particles are rejected by photophoresis and the deposition rate decreases. For very large radiative fluxes, photophoresis precludes the capture of char particles larger than a given size.

It is evident from these illustrative examples that the combination of photophoresis and thermophoresis induces a change in the size distribution in the mainstream through the dependence of α and β on particle size. By an appropriate combination of radiative and conductive fluxes, particle sizes larger than a given value can be avoided in the deposit and, for particles which present an extremal in the function $F(a/l)$ (as it is the case for fly ash particles) only a narrow width of particle sizes can be selected to deposit.

References

Castillo J.L., & Rosner D.E. (1988), "Theory of Surface Deposition from a Unary Dilute Vapor Containing Stream Allowing for Condensation within the Laminar Boundary Layer", *Chemical Engineering Science* (in press)

Goren S.L. (1977), "Thermophoresis of Aerosol Particles into Laminar Boundary Layer on a Flat Plate", *J. Colloid Interface Science* **61**, pp.77-85

Mackowski D.W. (1988), "Photophoresis of Aerosol Particles in the Free Molecular and Slip-Flow Regimes", *Int'l J. Heat Mass Transfer* (in press)

Rosner D.E. (1980), "Thermal (Soret) Diffusion Effects on Interfacial Mass Transport Rates", *J. PhysicoChemical Hydrodynamics* **1**, pp.159-185

Rosner D.E. (1985), "Mass Transfer across Gas Thermal Boundary Layers — Power Production and Materials Processing Implications", in **Heat Transfer in Fire and Combustion Systems**, HTD 45 (Law C.K. *et al.* eds.) ASME, N.Y.-N.Y., pp.3-8

Rosner D.E. & Kim S.S. (1984), "Optical Experiments on Thermophoretically Augmented Submicron Particle Deposition from 'Dusty' High Temperature Gas Flows", *The Chemical Engineering J.***29**, No.3, pp.147-157

Schlichting H. (1968), **Boundary Layer Theory,** McGraw-Hill (6th ed.)

Spalding D.B. & Evans M.L. (1961), "Mass Transfer through Laminar Boundary Layers — 3", *Int'l J. Heat Mass Transfer* **2**, pp.314-341

Talbot L., Cheng R.K., Schefer R.W. & Willis D.R. (1980), "Thermophoresis of Particles in a Heated Boundary Layer", *J. Fluid Mechanics* **101**, pp.737-758

Waldmann L. (1961), in **Rarefied Gas Dynamics** (Talbot L., ed.) Academic Press p.323

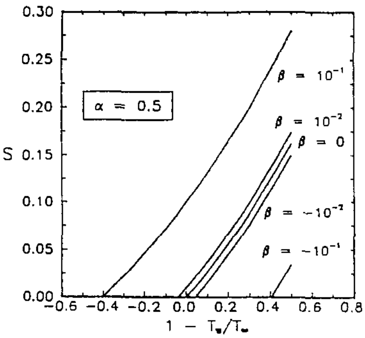

Figure 1. Dimensionless deposition rates S of particles as a function of $1-T_w/T_\infty$, for a constant thermophoretic coefficient $\alpha=0.5$ and different values of the photophoretic coefficient β.

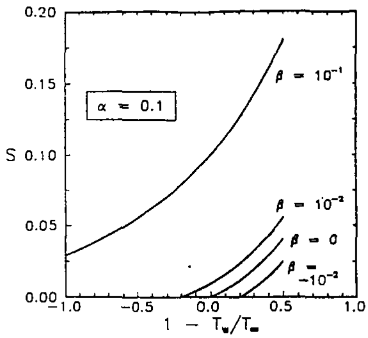

Figure 2. Dimensionless deposition rates S of particles as a function of $1-T_w/T_\infty$, for a constant thermophoretic coefficient $\alpha=0.1$ and different values of the photophoretic coefficient β.

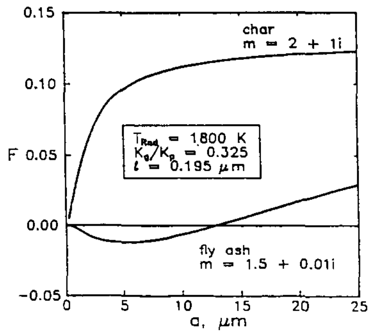

Figure 3. Photophoretic to thermophoretic velocity ratio for equal radiative and conductive heat flux, F, for carbonaceous char and fly-ash particles, as a function of particle radius. Radiation temperature $T_{Rad}=1800K$.

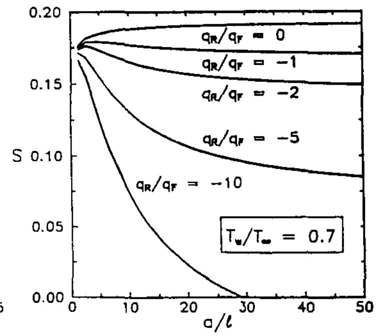

Figure 4. Dimensionless deposition rates S of char particles as a function of a/l, for $T_w/T_\infty=0.7$ and different ratios of the radiative to conductive heat flux at the surface.