Total Mass Deposition Rates from "Polydispersed" Aerosols

Daniel E. Rosner
High Temperature Chemical Reaction
Engineering Laboratory
Department of Chemical Engineering
Yale University
New Haven, CT 06520

Research in the last two decades has led to a greatly improved understanding of the nature of particle size *distributions* resulting from the various coagulation mechanisms (see, for example, the review of Friedlander, 1977, in Chapter 7), as well as of the laws governing single-sized particle deposition across gaseous boundary layers Rosner (1986). Quantitative information from both of these areas is, of course, needed to predict total mass deposition rates from "polydispersed" aerosols in engineering applications. In the present paper we demonstrate that, over a rather wide range of important conditions, total mass deposition rates from polydispersed aerosols resulting from coagulation, can be readily calculated from the corresponding deposition rate from a hypothetical "monodispersed" aerosol in which all particles have the prevailing average size (volume) $\bar{v}(=\phi_p/N_p)$. We illustrate our approach for the frequently encountered case of high-Peclet-number convective diffusion across nearly isothermal boundary layers, obtaining results that are remarkably insensitive to fluid dynamic conditions [laminar or turbulent boundary layer (BL)], particle Knudsen number (free-molecule or continuum Brownian diffusion) or particle morphology (dense spherical particles or low-density agglomerates). Generalizations with respect to particle deposition mechanism and shape of the particle size distribution are then indicated, along with potential engineering applications.

Convective-Diffusion Mass Deposition Rates from Particle Size Distributions

Suppose the particle size distribution at the outer edge of a gaseous boundary layer is $n\{v\}$; defined such that the total particle mass fraction is:

$$\omega_p = \frac{1}{\rho} \cdot \int_0^\infty \tilde{\rho}_p\{v\} \cdot v \cdot n\{v\}\, dv \qquad (1)$$

where v is the individual particle volume (treated here as a continuous variable), ρ is the mixture density and $\tilde{\rho}_p\{v\}$ is the intrinsic density of a particle of volume v. If the local dimensionless mass transfer coefficient (Stanton number) is written $St_m\{v, \ldots\}$, then the total mass deposition rate can be formally written:

$$-\dot{m}_p'' = U \cdot \int_0^\infty St_m\{v, \ldots\} \cdot \tilde{\rho}_p\{v\} \cdot v \cdot n\{v\}\, dv \qquad (2)$$

showing that $-\dot{m}_p''$ is proportional to the integral of the particle size distribution function after "weighting" by the function: $St_m\{v, \ldots\} \cdot \tilde{\rho}_p\{v\} \cdot v$. In the cases examined explicitly, Figure 1, we assume:

1. $St_m \sim Sc^{-b}$ where, for high Peclet number transport, $b = 2/3$ for laminar BLs, and 0.701 for turbulent BLs (Friedlander, 1977; Rosner, 1986). [High Pe behavior even applies to *low* Reynolds number flows because of the largeness of the particle Schmidt number (smallness of the particle Brownian diffusivity compared to the host gas momentum diffusivity, ν) (Friedlander, 1977; Fernandez de la Mora and Rosner, 1982).]
2. $n\{v\}$ is "self-preserving" in the sense that $n\bar{v}/N_p = \Psi$ is a calculable function of $v/\bar{v} \equiv \eta$ (Friedlander, 1977; Friedlander and Wang, 1966; Lai *et al.*, 1977)
3. $\tilde{\rho}_p\{v\}$ is a constant independent of v
4. Further coagulation within the fluid-dynamic boundary layer is neglected (for a discussion of coagulation in thermophoretically-dominated situations, see Park and Rosner, 1988)

Here Sc, the particle Schmidt number, is the diffusivity ratio ν/D_p, where D_p is the particle Brownian diffusivity in the prevailing gas; and $\bar{v}$ is the mean particle volume defined by ϕ_p/N_p, where:

$$\phi_p \equiv \int_0^\infty v n\{v\}\, dv \quad \text{(total particle volume fraction)} \qquad (3)$$

$$N_p \equiv \int_0^\infty n\{v\}\, dv \quad \text{(total particle number density)} \qquad (4)$$

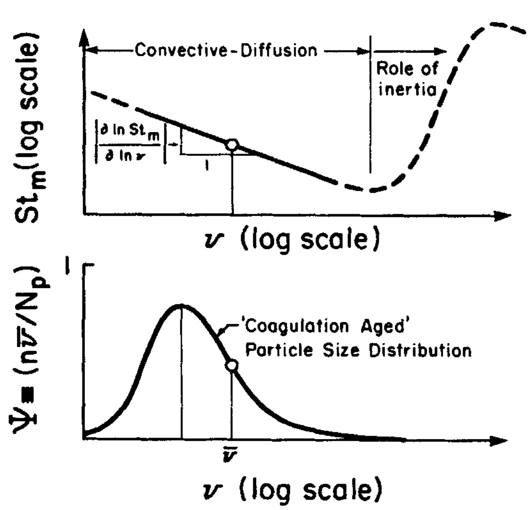

Figure 1. Behavior of the dimensionless mass transfer coefficient, St_m, and dimensionless aerosol particle size distribution, Ψ, as a function of the logarithm of the particle volume.

The purpose of this note is to compare the total mass deposition rate computed from Eq. 2 with the appropriate *reference* value:

$$-\dot{m}''_{p,\text{ref}} = U \cdot St_m(\bar{v}) \cdot \tilde{\rho}_p(\bar{v}) \cdot \bar{v} N_p \tag{5}$$

or, the corresponding mass deposition rate from a hypothetical "monodispersed" aerosol comprised particles each having the volume $\bar{v}$. [For the special case of $Sc \gg 1$ deposition of dense spherical particles from a turbulent dusty gas flow in a circular duct, Friedlander (1977) has made a similar type of calculation, but using a rather different value of $-\dot{m}''_{p,\text{ref}}$, based on the deposition rate of the "monomer" (vapor) from which the particles were presumably formed, a choice which is not of the same order of magnitude as $-\dot{m}''_{p,\text{actual}}$.]

For dense spheres, two limiting cases are of special interest, *viz.* Knudsen ("free-molecule") diffusion, in which case:

$$D_p \sim v^{-2/3} \tag{6}$$

and Stokes-Einstein ("continuum") diffusion, in which case:

$$D_p \sim v^{-1/3} \tag{7}$$

For quasispherical agglomerates comprised primary particles which sum to v, the interesting recent work of Mountain, Mulholland, and Baum (1986) indicates that Eqs. 6 and 7 generalize these proportionalities to:

$$D_p \sim v^{-2/D} \quad \text{(Knudsen)} \tag{8}$$

and:

$$D_p \sim v^{-1/D} \quad \text{(Stokes-Einstein)} \tag{9}$$

where the "fractal" dimension (near 1.8) replaces 3 in the indicated exponents. Combining these laws reveals that in each of the abovementioned cases:

$$\frac{-\dot{m}''_{p,\text{actual}}}{-\dot{m}''_{p,\text{ref}}} = \int_0^\infty \eta^k \cdot \Psi(\eta) d\eta \equiv \mu_k \tag{10}$$

for some appropriate value of k calculated from the exponent b, the diffusion regime, and the fractal "dimension" D. Accordingly, calculation of the total mass deposition rate from such polydispersed aerosols reduces to a computation of $-\dot{m}''_{p,\text{ref}}$ (based on all particles of size $\bar{v}$), corrected only by an appropriate (dimensionless) "moment" of the self-preserving size distribution function. The required correction factors are readily estimated below by interpolating between the selected moments numerically computed earlier by Wang and Friedlander (1967), and Lai, Friedlander, Pich and Hidy (1972). It should be remarked that, as a consequence of the definitions of Ψ and η and Eqs. 3 and 4, both the zeroth moment and first moment of $\Psi(\eta)$ must be unity.

Required "Moments" for Convective Diffusion

Table 1 collects the resulting values of the exponent k in Eq. 10 for each of the particle deposition cases considered here, as well as the corresponding estimated moments μ_k, Figure 2. It is interesting to note that these k-values fall between 0.218 and 0.778 and, as shown in Figure 2, between $k = 0$ and 1, both Knudsen-$\Psi(\eta)$ and the continuum-$\Psi(\eta)$functions have moments which exhibit a very shallow minimum near $k = 1/2$. [Until reliable $\Psi(\eta)$ and μ_k-values become available for the two $D = 1.8$ cases, these particular estimates (especially for the Knudsen diffusion case) should be viewed as tentative.] As a consequence of this property, and of the abovementioned requirements $\mu_0 = 1$, $\mu_1 = 1$, all of the inferred μ_k-values (cf. Eq. 10) fall in a remarkably narrow band between 0.88 and 0.92. This leads to a

Table 1. Parameter Values, Required Moments, and Predicted Ratios, of $-\dot{m}''_p$ (Polydispersed)/$-\dot{m}''_p$ (Monodispersed with $v = \bar{v}$) for High-Peclet-Number Convective Diffusion of Dense Particles and Agglomerates from Coagulation-Aged Aerosols

Case		Knudsen Diffusion		Continuum Diffusion	
Sc-Exponent (b)	Fractal Dimension (D)*	Relevant Moment (k)	Moment Value (μ_k)	Relevant Moment (k)	Moment Value (μ_k)
2/3 (LBL)	3 (dense)	0.5556	0.888	0.7778	0.924
2/3 (LBL)	1.8 (agglom.)	0.2593	0.901*	0.6296	0.901*
0.704 (TBL)	3 (dense)	0.5307	0.902	0.7653	0.922
0.704 (TBL)	1.8 (agglom.)	0.2178	0.919*	0.6089	0.899*

*Values of μ_k for the $D = 1.8$ (fractal agglomerate cases) should be considered tentative estimates (especially for Knudsen diffusion) pending numerical calculations of $\Psi(\eta)$ for the relevant collision kernels.

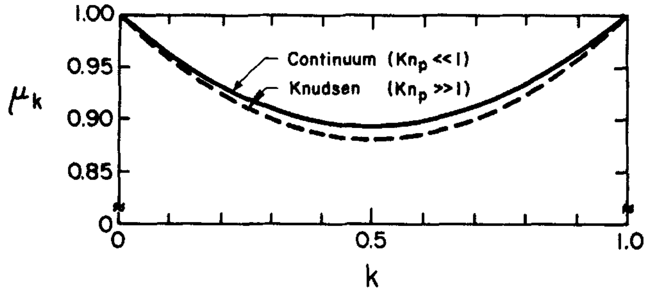

Figure 2. Behavior of the moments of the self-preserving size distribution functions $\Psi(\eta)$ for Knudsen diffusion ($Kn_p \gg 1$, dashed line) and continuum diffusion ($Kn_p \ll 1$) for k values between zero and unity.
Compare with Eq. 2, ($k = 1 + (\partial \ln St_m/\partial \ln v)$.

simplification of potential conceptual and engineering utility—namely, *to a first approximation, total mass deposition rates by convective-diffusion from "coagulation-aged" aerosol population distributions will be about 90% of the values corresponding to a hypothetical "monodispersed" aerosol* [*with all particles having the mean particle size (volume)* $\bar{v}(\equiv \phi_p/N_p)$] *under otherwise identical fluid dynamic and geometric conditions.* To within about ±2%, these results are insensitive to the fluid dynamic state of the boundary layer, particle Knudsen number (ratio of gas mean free path to particle diameter) and, evidently, even apply to "open" agglomerates.

Generalizations and Implications

It can be seen from the abovementioned formulation that the following conditions will lead to $-\dot{m}_p''/-\dot{m}_{p,\text{ref}}''$ ratios *even closer to unity:*

1. The simultaneous role of particle *thermophoresis,* which is known to reduce the absolute value of $\partial \ln St_m/\partial \ln v$ when the target is cooler than the gas stream (see Rosner, 1980; 1986)
2. Mainstream particle size distributions which are *narrower* than those corresponding to coagulation-aged aerosols, *i.e.,* narrower than the "self-preserving" distributions $\Psi(\eta)$, or their near-"log-normal" equivalents (with $\sigma_g \approx 1.32$–1.35) (Lee, 1983; Lee *et al.,* 1984)

Of course, in general, $St_m(v)$ is not a simple power law over the *entire* particle size range, Figure 1, but we can verify that a "local" power-law approximation is adequate by expanding $\ln St_m$ *vs* $\ln v$ in a Taylor series about $\ln \bar{v}$, *i.e.:*

$$\ln St_m \simeq (\ln St_m)_{v=\bar{v}} + \left(\frac{\partial \ln St_m}{\partial \ln v}\right)_{v=\bar{v}} \cdot \ln\left(\frac{v}{\bar{v}}\right) + \frac{1}{2!}\left(\frac{\partial^2 \ln St_m}{\partial \ln v^2}\right)_{v=\bar{v}} \cdot \left(\ln \frac{v}{\bar{v}}\right)^2 + \cdots \quad (11)$$

Inserting Eq. 11 into Eq. 2 and introducing the definitions of Ψ and η, one can show that the power-law approximation is adequate provided:

$$\left|\frac{1}{2} \cdot \left(\frac{\partial^2 \ln St_m}{\partial \ln v^2}\right)_{v=\bar{v}}\right| \cdot \frac{1}{\mu_k} \cdot \int_0^\infty (\ln \eta)^2 \cdot \eta^k \cdot \Psi(\eta) \cdot d\eta \ll 1 \quad (12)$$

where, as before:

$$k = 1 + \left(\frac{\partial \ln St_m}{\partial \ln v}\right)_{v=\bar{v}} \quad (13)$$

and μ_k is defined by Eq. 10.

It is known that the value of $(\partial \ln St_m/\partial \ln v)_{v=\bar{v}}$ will increase with the onset of particle "inertia" effects at the larger particle sizes. Of course, this will increase μ_k and, ultimately, drive $-\dot{m}_p''/-\dot{m}_{p,\text{ref}}''$ above unity. For example, the phenomenon called "eddy impaction" causes $St_m/(C_f/2)^{1/2}$ for turbulent flow through smooth circular ducts to increase like $(t_p^+)^2$ over an approximately 2.5 decade range of t_p^+, (see, e.g., Papavergos and Hedley, 1984), where:

$$t_p^+ \equiv \frac{u_*^2 t_p}{\nu} \quad (14)$$

Here u_* is the "friction velocity" $(\bar{\tau}_w/\rho)^{1/2}$, and t_p the characteristic particle "stopping" time. Under continuum ($Kn_p \ll 1$) conditions, $t_p \sim d_p^2 \sim v^{2/3}$ so that, over nearly a 2-decade range of v, this corresponds to:

$$\frac{\partial \ln St_m}{\partial \ln v} = +\frac{4}{3} \quad (15)$$

and hence, $k = 7/3 = 2.333$. Interpolating, using values of μ_2 and μ_3 reported earlier by Wang and Friedlander (1967), we find $\mu_{7/3} \approx 2.9$ under such "eddy-impaction" conditions. While it is true that jet impaction situations (as in a "cascade impactor") can lead to still larger local values of $\partial \ln St_m/\partial \ln v$, for most external- and internal-flow situations μ_k will fall between the abovementioned limits of *ca.* 0.9 (convective diffusion) and *ca.* 2.9 (eddy impaction-modified turbulent transport). Accordingly, $-\dot{m}_p''/-\dot{m}_{p,\text{ref}}''$ will normally fall between these same limits for a wide class of more general $St_m(v)$ functions, *including the now-classical "capture efficiency" correlations* for steady inviscid flow past isolated cylinders or spheres, (Rosner, 1986; Israel and Rosner, 1983; Friedlander, 1977).

Acknowledgments

It is a pleasure to acknowledge the financial support of DOE-PETC *via* Grant No. DE-FG22-86PC90756 and the US AFOSR *via* Grant No. AFOSR-84-0034, as well as the related support of the Yale HTCRE Laboratory by our current industrial affiliates: Shell Foundation, AVCO-Lycoming-Textron, and SCM-Chemicals. In this research, the author has benefited from helpful discussions with Professors J. Fernandez de la Mora and J. O'Brien, as well as his current and former graduate students.

The author wishes to dedicate this paper to the memory of aerosol dynamics researcher, Prof. Isaiah Gallily (Hebrew U.), who died on July 31, 1988.

Literature Cited

Fernandez de la Mora, J., and D. E. Rosner, "Effects of Inertia on the Diffusional Deposition of Small Particles to Spheres and Cylinders at Low Reynolds Numbers." *J. Fluid Mechanics,* **125,** 379 (1982).

Friedlander, S. K., *Smoke, Dust and Haze-Fundamentals of Aerosol Behavior,* Wiley, New York (1977).

Friedlander, S. K., and C. S. Wang, "The Self-Preserving Particle Size Distribution for Coagulation by Brownian Motion," *J. Colloid & Interf. Sci.,* **22,** 126 (1966).

Lai, F. S., S. K. Friedlander, J. Pich, and G. M. Hidy, "The Self-Preserving Particle Size Distribution for Brownian Coagulation in the

Free-Molecule Regime," *J. Colloid. & Interf. Sci.*, **39**(2), 395 (May 1972).

Lee, K. W., "Change of Particle Size Distribution during Brownian Coagulation," *J. Colloid & Interf. Sci.*, **92**, 315 (1983).

Lee, K. W., H. Chen, and J. A. Gieseke, "Log-Normal Preserving Size Distribution for Brownian Coagulation in the Free-Molecule Regime," *Aerosol Sci. & Tech.*, **3**, 53 (1984).

Mountain, R. D., G. W. Mulholland, and H. Baum, "Simulation of Aerosol Agglomeration in Free-Molecular and Continuum Flow Regimes," *J. Colloid & Interf. Sci.*, **114**(1), 67 (1986).

Papavergos, P. G., and A. B. Hedley, "Particle Deposition Behavior from Turbulent Flows," *Chem. Eng. Res. Dev.*, British Inst. Chem. Engs., **62**, 275 (1984).

Park, H. M., and D. E. Rosner, "Effect of Coagulation in the Boundary Layer on the Size Distribution of Thermophoretically Deposited Particles," *Chem. Eng. Sci.* (in press); Appendix 5 of D. E. Rosner and H. M. Park, "Thermophoretically Augmented Mass-, Momentum- and Energy-Transfer Rates in High Particle Mass-Loaded Laminar Forced Convection Systems," *Chem. Eng. Sci.*, **43**, 2689 (1988).

Rosner, D. E., "Thermal (Soret) Diffusion Effects on Interfacial Mass Transport Rates," *J. Phys. Chem. Hydrody.*, **1**, 159 (1980).

Rosner, D. E., *Transport Processes in Chemically Reacting Flow Systems*, Butterworth, Stoneham, MA (1986); second printing, (1988).

Wang, C. S., and S. K. Friedlander, "The Self-Preserving Particle Size Distribution for Coagulation by Brownian Motion," *J. Colloid & Interf. Sci.*, **24**, 170 (1967).

Manuscript received May 2, 1988, and revision received Aug. 9, 1988.