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EFFECT OF DIFFUSIOPHORESIS ON PARTICLE COLLECTION BY WET SCRUBBERS

LESLIE E. SPARKS and MICHAEL J. PILAT

Water and Air Resources Division, Department of Civil Engineering, University of Washington, Seattle, Washington

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Abstract—The effects of diffusiophoresis on particulate collection by wet scrubbers are considered. Single droplet collection efficiencies are calculated for particle collection by the combined mechanisms of inertial impaction and diffusiophoresis. The resulting single droplet particle collection efficiencies are used to calculate overall scrubber efficiencies for three cases, positive, negative and zero diffusiophoresis. The calculations indicate that negative diffusiophoresis (droplet evaporation) can cause poor scrubber performance, especially for small particles, whereas positive diffusiophoresis (condensation) can result in greatly improved particulate collection.

Nomenclature

A_x = x component of nondimensional particle acceleration due to diffusiophoresis
A_y = y component
a = dimensional particle acceleration vector
C = concentration of scrubbing liquid vapor
D_{vg} = diffusivity of scrubbing liquid vapor in gas
dp/dx = vapor pressure gradient
E_c = collision efficiency
E_d = particle collection efficiency of a single droplet
E_0 = scrubber overall particle collection efficiency
F_D = force due to diffusiophoresis
F_d = drag force
F_e = vector sum of all external forces
f = fraction of gas swept by liquid droplets
G = gas flow rate
H = distance traveled by droplet with respect to the gas
K = Stokes number
Kn = Knudsen number
L = liquid flow rate
m = particle mass
m_g = molecular mass of gas
m_v = molecular mass of vapor
N = total number of particles impinging on droplet
N_c = number of particles collected
n = total number of gaseous molecules per unit volume
p_g = partial pressure of the carrier gas
p_B = partial pressure of scrubbing liquid vapor in gas bulk phase
p_0 = saturation vapor pressure of scrubbing liquid
R = droplet radius
r = particle radius
T = nondimensional time
t = dimensional time
U_s = velocity of gas due to Stephan flow
U_X = nondimensional X component of fluid velocity
U_Y = nondimensional Y component of fluid velocity
u_0 = undisturbed fluid velocity (dimensional)
u_x = dimensional x component of fluid velocity
u_y = dimensional y component of fluid velocity
V_x = nondimensional X component of particle velocity
V_y = nondimensional Y component of particle velocity

v_m = particle velocity due to gas momentum transfer
v_p = particle velocity due to diffusiophoresis
v_s = particle velocity due to Stephan flow
v_x = dimensional x component of particle velocity
v_l = dimensional y component of particle velocity
X = nondimensional X coordinate
x = dimensional x coordinate
Y = nondimensional y coordinate
Y_0 = initial Y position of grazing trajectory
y = dimensional y coordinate
y_0 = initial y position of grazing trajectory

Greek letters

γ_g = molar fraction of gas
γ_v = molar fraction of vapor
λ = gas mean free path
μ = viscosity of gas
ρ = density of gas-vapor mixture
ρ_p = particle density
τ = particle relaxation time

1. INTRODUCTION

(a) *Lack of theory to predict scrubber performance*

ALTHOUGH scrubbers are commonly used to remove particulate matter from effluent gas streams, there are few theoretical equations relating scrubber performances to scrubber design variables and operating parameters. The lack of such knowledge hinders improvement in the design and operation of scrubbers and makes comparisons between the performance data reported in the literature difficult.

KLEINSCHMIDT (1939) has shown that the overall scrubber particulate collection efficiency E_0 can be calculated by

$$E_0 = 1 - e^{-fE_d} \tag{1}$$

Where f is the fraction of gas swept by liquid droplets

$$f = \frac{3\,H\,L}{4\,R\,G} \tag{2}$$

H is the distance the droplet travels with respect to gas, L/G is the liquid to gas flow ratio, R the droplet radius and E_d the particle collection efficiency of a single droplet. The single droplet collection efficiency is a function of the various forces acting on the particle (inertial, Brownian diffusion, electrostatic, diffusiophoresis, etc.).

This paper presents theoretical calculations of the single droplet collection efficiency for particles subjected to inertial and diffusiophoresis forces. The resulting single droplet collection efficiencies are then used in equation (1) to calculate the overall scrubber efficiency.

(b) *Reports on apparent diffusiophoresis effects*

SEMRAU *et al.* (1955) reported that the observed performance differences between particle collection by a pipeline scrubber and a Venturi–cyclonic spray scrubber were probably due to differences in the scrubbing liquid temperature. LAPPLE and KAMACK (1955) reported that the addition of steam (2–3 times that necessary to saturate the air at room temperature) produced a fivefold reduction in dust loss at a given air pressure drop. They attributed the increased particulate collection to vapor diffusion and steam condensation mechanisms. SCHAUER (1955) reported that 99.9 per cent removal of DOP smoke was obtained with a steam nozzle (venturi) Pease–Anthony scrubber using a steam pressure of 48 psig, 0.2 lb steam/1000 ft^3 gas, 13 gal scrubbing liquid/1000 ft^3 gas, and 0.68 gal cooling water/1000 ft^3 gas. The high particulate removal efficiency was thought to be due to diffusion–impaction mechanisms.

(c) *Causes of diffusiophoresis in scrubbers*

The scrubbing liquid equilibrium vapor pressure and the concentration of scrubbing liquid vapor in the gaseous phase (humidity in a water–air scrubber) determine the scrubbing liquid vapor pressure gradient. The liquid vapor pressure gradient is positive for an evaporating droplet and negative for a droplet upon which vapor is condensing. Whenever the liquid vapor pressure gradient is finite, diffusiophoresis is present in the scrubber. Diffusiophoresis may either aid or hinder the collection of particles by droplets. The diffusiophoresis force is defined as positive if particle collection is aided and negative if particle collection is hindered.

The scrubbing liquid vapor pressure gradient may be described by

$$\frac{\mathrm{d}p}{\mathrm{d}x} = \frac{\Delta p}{\Delta x} = \frac{p_0 - p_B}{\Delta x} \tag{3}$$

where p_0 is the saturation vapor pressure of the scrubbing liquid, p_B is the partial pressure of the scrubbing liquid vapor in the gas bulk phase, and Δx is the distance necessary to go from the saturation vapor pressure to the gas bulk phase vapor pressure of the scrubbing liquid.

Conditions conducive to vapor pressure gradients may also result in thermal gradients. These thermal gradients impose an additional force, called thermophoresis, on the particles. Calculations reported by HORST (1968) indicate that the force due to thermophoresis may be much less than the force due to diffusiophoresis for many situations in an air stream mixture. Therefore, the force due to thermophoresis is neglected in the following calculations.

2. DIFFUSIOPHORESIS THEORY

(a) *Stephan flow*

The diffusiophoresis force on a particle is the sum of the force due to Stephan flow and a force due to gas momentum transfer processes. Stephan flow is the hydrodynamic flow necessary to maintain a uniform total pressure in a diffusing gaseous system and is directed away from a surface where liquid is evaporating and towards a surface where vapor is condensing. The existence of such a flow was first postulated by STEPHAN (1881) and later verified by FACY (1957). In a binary system of a vapor and a carrier gas the velocity of the Stephan flow is

$$U_s = -\frac{D_{vg}}{p_g}\frac{\mathrm{d}p}{\mathrm{d}x} \tag{4}$$

where D_{vg} is the diffusion coefficient of the vapor in the gas and p and p_g are the partial pressures of the vapor and carrier gas, respectively. If a particle near a surface where evaporation or condensation is occurring moves with the Stephan velocity (valid for small particles), the particle velocity due to Stephan flow is

$$v_s = U_s = -\frac{D_{vg}}{p_g}\frac{dp}{dx} \tag{5}$$

For an air–water system at 0°C and 1 atm total pressure the particle velocity due to Stephan flow is

$$v_s = -2.4 \times 10^{-4}\frac{dp}{dx} \tag{6}$$

where dp/dx is in mbar/cm.

(b) *Particle radius smaller than gas mean free path*

If the molecular masses of the diffusing gas molecules are different, the motion of small particles (particle radius smaller than the gas mean free path) in the diffusing mixture of gases is affected by gas momentum transfer processes. In this situation the particle no longer moves with the Stephan velocity.

BAKANOV and DERGAGUIN (1957, 1960) and WALDMANN (1959) have derived an equation for the particle velocity based on a rigorous consideration of the effects of diffusing gas molecules on particle motion using the Chapman–Enskog theory of gases. The particle velocity caused by the collisions with diffusing gas molecules is given by

$$v_m = -\frac{\sqrt{m_v} - \sqrt{m_g}}{\gamma_v\sqrt{m_v} + \gamma_g\sqrt{m_g}} D_{vg}\frac{d\gamma_v}{dx} \tag{7}$$

where m_v, m_g, γ_v and γ_g are the mass and molar fraction of the vapor and gas, respectively.

As shown by GOLDSMITH and MAY (1966) and GOLDSMITH *et al.* (1963) the particle total velocity (diffusiophoresis velocity) is the sum of the Stephan flow velocity and the velocity due to gas–particle momentum transfer. In a dilute vapor system ($p_v \ll p_g$) the particle total velocity is

$$v_p = \frac{-\sqrt{m_v}}{\gamma_v\sqrt{m_v} + \gamma_g\sqrt{m_g}}\frac{D_{vg}}{p_g}\frac{dp}{dx}. \tag{8}$$

For water vapor diffusing through air at 0°C and 1 atm total pressure the particle diffusiophoresis velocity given by equation (8) is

$$v_p = -1.9 \times 10^{-4}\frac{dp}{dx} \tag{9}$$

which compares well with the measurements reported by GOLDSMITH *et al.* (1963) given by

$$v_p = -1.89 \times 10^{-4}\frac{dp}{dx} \tag{10}$$

(c) *Particle radius larger than gas mean free path*

The assumptions concerning the gas flow field around the particle used to derive equation (8) are not valid for particles larger than the gas mean free path λ. SCHMIDT and WALDMANN (1960) reported that the particle velocity predicted by equation (8) differs from the true particle velocity by about 10 per cent.

DERGAGUIN *et al.* (1966) have extended the theory of particle motion in a diffusing binary gas system to account for the new flow regime. Their equation for the particle velocity is

$$v_p = \frac{D_{vg}\, n(m_p - m_v)}{\rho} \frac{\Delta C}{\Delta X} \tag{11}$$

where n is the total number of gaseous molecules per unit volume, $\Delta C/\Delta X$ is the vapor concentration gradient and ρ is the density of the gas–vapor mixture. DERJAGUIN *et al.* (1966) reported experimental results which show that equation 11 holds for Knudsen numbers ($Kn = \lambda/r$) less than 0.5 and that equation (8) is valid for Knudsen numbers greater than 0.7.

3. COLLECTION OF PARTICLES BY DROPLETS

(a) *Inertial impaction*

The single droplet collection efficiency due to inertial impaction is given by

$$E_a = \frac{y_0^{\,2} E_c}{R^2} \tag{12}$$

where y_0 is the initial y position, measured from the drop centerline, of the particle trajectory that just grazes the droplet, R is the radius of the droplet and E_c is the collision efficiency

$$E_c = \frac{N_c}{N} \tag{13}$$

where N_c is the number of particles collected by the droplet and N is the total number of particles impinging upon the collector droplet. The collision efficiency E_c is generally assumed to be unity.

Diffusiophoresis can influence the single droplet collection efficiency by changing the particle trajectory and/or by modifying the particle collision efficiency E_c.

(b) *Effect of diffusiophoresis on single droplet collection*

Efficiency by inertial impaction. The effect of diffusiophoresis on the trajectories of particles flowing near droplets can be determined by solving the equation of particle motion for a gas flowing around a sphere. The equation of particle motion is developed in Appendix I. The X and Y components of the nondimensional equation of particle motion are:

$$\frac{d^2X}{dT^2} = \frac{1}{K}(U_x - V_x) + A_x \tag{14}$$

$$\frac{d^2Y}{dT^2} = \frac{1}{K}(U_y - V_y) + A_y \tag{15}$$

where X and Y are nondimensional distances, T is nondimensional time, K the Stokes number ($K = 2r^2\rho_p u_0/\mu R$), r the particle radius, ρ_p the particle density, u_0 the undisturbed fluid velocity, U_x and U_y the nondimensional fluid velocity components,

V_x and V_y the nondimensional particle velocity components and A_x and A_y the nondimensional components of the particle acceleration caused by diffusiophoresis.

The solution of equations (14) and (15) requires information concerning the gas flow field U_x and U_y. For potential flow (gas considered to be inviscid, irrotational and incompressible) U_x and U_y are given by

$$U_x = 1 - \frac{2X^2 - Y^2}{2(X^2 + Y^2)^{2 \cdot 5}} \tag{16}$$

$$U_y = - \frac{3XY}{2(X^2 + Y^2)^{2 \cdot 5}}. \tag{17}$$

The single droplet collection efficiency in terms of the nondimensional parameters can be obtained by substituting $y = YR$ into equation (12) to give

$$E_d = Y_0{}^2 E_c. \tag{18}$$

The magnitude of Y_0 is determined by solving equations (14) and (15) for the particle trajectory that just grazes the droplet. The initial conditions used in the Runge–Kutta numerical solution are at $T = 0$, $X = -5.0$ and $Y = Y^0$.

The calculated single droplet particle collection efficiencies for condensation, evaporation and no gas–liquid mass transfer are shown in FIG. 1 for a collision

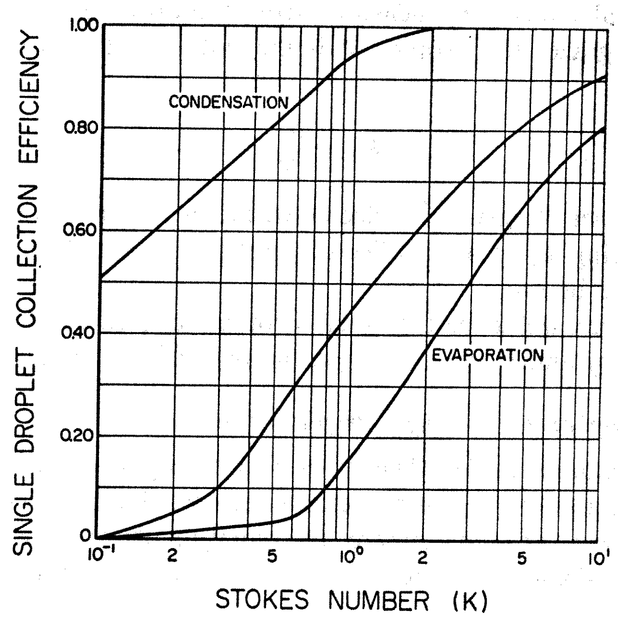

FIG. 1. Effect of diffusiophoresis on single droplet collection efficiency.

efficiency E_c equal to unity. The upper curve is for water condensation ($dp/dx = -10^5$ mbar/cm), the middle curve is for no water gas–liquid mass transfer ($dp/dx = 0$), and the lower curve is for water evaporation ($dp/dx = 10^5$ mbar/cm). The undisturbed gas velocity u_0 was 100 cm/s for all three cases.

The effect of diffusiophoresis on particle collection by inertial impaction is greater for the smaller particles. For example the difference in the particle collection efficiency by a 100 μ radius droplet moving through air at 100 cm/s between condensation and evaporation at a vapor pressure gradient of 500 mbar/cm is an order of magnitude for a 1 μ radius particle but less than 1 per cent for an 18 μ radius particle.

In general the effect of diffusiophoresis decreases as the relative velocity of the droplet with respect to the gas increases. This is in agreement with the experimental results reported by TOVBIN *et al.* (1965).

(d) *Effect of diffusiophoresis on collision efficiency*

The effect of diffusiophoresis on the particle collision efficiencies is essentially unknown. A quantitative prediction of the collision efficiency is presently impossible and the reported experimental observations are contradictory. The results reported by PROKHOROV (1954) indicate that the collision efficiency may be less than unity in the presence of negative diffusiophoresis (evaporation). However, TOVBIN *et al.* (1965) have reported that at high particle Reynolds numbers the collision efficiency is unity regardless of vapor pressure gradients.

4. OVERALL SCRUBBER EFFICIENCY

The single droplet efficiencies from FIG. 1 can be used with equation (1) to calculate overall scrubber efficiencies for positive, negative and zero diffusiophoresis. The results of these calculations for particle collection in a scrubber with a droplet travel H of 300 cm (about 10 ft); a liquid to gas flow rate ratio L/G of 5×10^{-4} (corresponds to 3.7 gal/1000 CFM); and a droplet radius R of 100 μ are presented in FIG. 2. As in FIG. 1 the upper curve is for condensation ($dp/dx = -10^5$ mbar/cm), the middle curve is for no mass transfer ($dp/dx = 0$), and the lower curve is for evaporation ($dp/dx = +10^5$ mbar/cm).

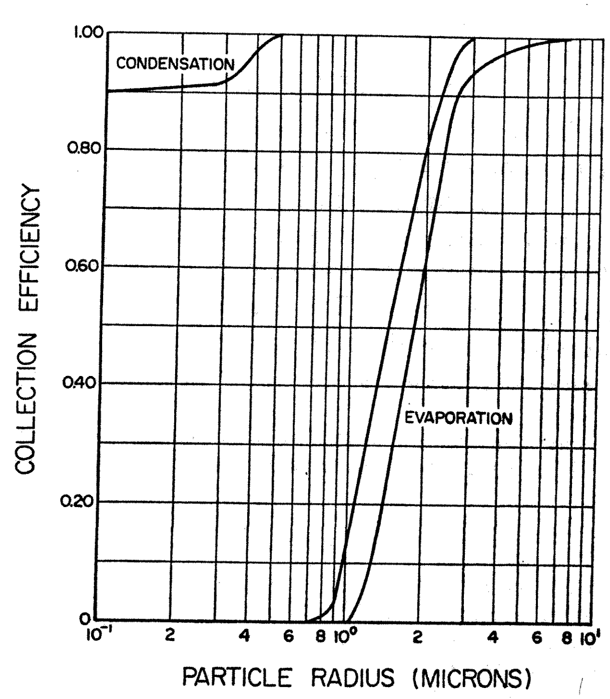

FIG. 2. Effect of diffusiophoresis on overall scrubber collection efficiency.

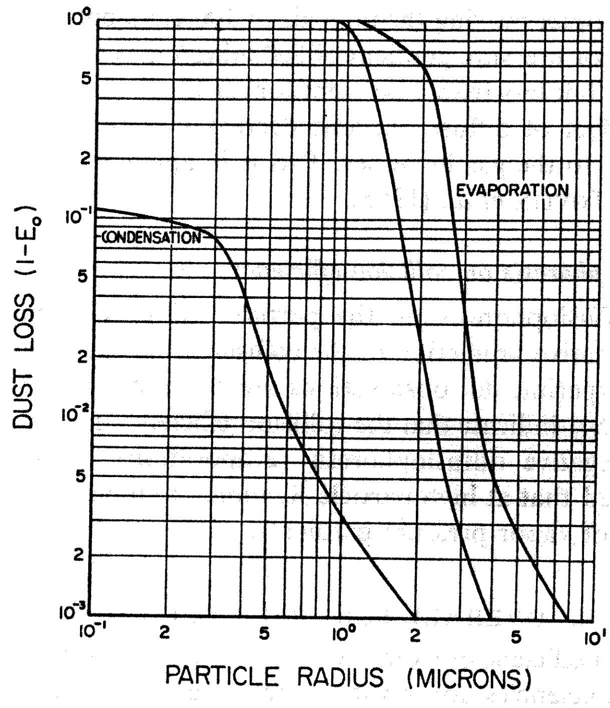

FIG. 3. Effect of diffusiophoresis on dust loss from a scrubber.

The effect of diffusiophoresis on the amount of particulate matter released to the atmosphere is shown in FIG. 3 where dust loss $(1 - E_0)$ is plotted vs. particle radius for the same three cases as in FIGS. 1 and 2. In this case the upper curve is for evaporation, the middle curve is for no mass transfer, and the lower curve is for vapor condensation. The mass of particulate emitted to the atmosphere is determined by multiplying the particulate mass flow rate by the dust loss.

5. CONCLUSIONS

The calculated results shown in FIGS. 1 and 2 plus the reported experimental observations indicate that the scrubbing liquid temperature and vapor concentration may play a major role in determining scrubber performance, especially for small particles. Additional information concerning scrubber liquid temperature, vapor concentration, droplet size distribution and particle collection efficiencies are needed in order to verify the calculated results. The need for this data is increasingly important in light of the proposed air quality standards for particulate air pollutants which will require particle removal efficiencies near 100% at emission sources.

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APPENDIX I

Development of equations of motion for a spherical particle past a spherical collector

From Newton's second law

$$m\mathbf{a} = \Sigma\mathbf{F}(t) \tag{19}$$

where m is the particle mass $=(4/3)\pi\rho_p r^3$, ρ_p is the particle density, r is the particle radius, a is the particle acceleration

$$\mathbf{a} = \mathrm{d}v/\mathrm{d}t \tag{20}$$

$\mathbf{v}$ is the particle velocity, t is time and $\Sigma\mathbf{F}(t)$ is the vector sum of all external forces acting on the particle, which may be a function of time. Assume that the only forces acting on the particle are the force due to the resistance of the medium to particle motion, i.e. a drag force, $\mathbf{F}_d$ and $\mathbf{F}_c(t)$ the vector of all other external forces. Further assume that the drag force is given by Stokes' law.

$$\mathbf{F}_d = 6\pi\mu r\,(\mathbf{u} - \mathbf{v}) \tag{21}$$

where μ is the viscosity of the fluid, and $\mathbf{u}$ is the fluid velocity. Substitute for $\Sigma\mathbf{F}(t)$ into equation (19) to get

$$m\mathrm{d}\mathbf{v}/\mathrm{d}t = 6\pi\mu r\,(\mathbf{u} - \mathbf{v}) + F_c(t) \tag{22}$$

Dividing equation (22) by m and defining a relaxation time, τ as

$$\tau = m/6\pi\mu r \tag{23}$$

or for spheres

$$\tau = 2\rho_p r^2/9\mu \tag{24}$$

we get

$$\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \frac{1}{\tau}(\mathbf{u} - \mathbf{v}) + \frac{\mathbf{F}_c(t)}{m}. \tag{25}$$

Assume that the only other external force is the force due to diffusiophoresis F_D given by

$$F_D = 6\pi\mu r v_p \tag{26}$$

where v_p is the particle velocity due to diffusiophoresis. Equation (25) becomes

$$\frac{d\mathbf{v}}{dt} = \frac{1}{\tau}(\mathbf{u} - \mathbf{v}) + \frac{F_D}{m}. \tag{27}$$

The x and y components of equation (27) are

$$\frac{dv_x}{dt} = \frac{1}{\tau}(u_x - v_x) + \frac{F_{D-x}}{m}. \tag{28}$$

and

$$\frac{dv_y}{dt} = \frac{1}{\tau}(u_y - v_y) + \frac{F_{D-y}}{m}. \tag{29}$$

Equations (28) and (29) are solved with the initial conditions that at $t = 0, x = -\infty, u_x = v_x = u_0$ where u_0 is the undisturbed fluid velocity and $u_y = v_y = 0$.

Equations (28) and (29) are converted to nondimensional form by making the following substitutions:

$$X = x/R \qquad Y = y/R$$

$$V_x = v_x/u_0 = dX/dT$$

$$V_y = v_y/u_0 = dY/dT$$

$$T = tu_0/R$$

$$K = 2\rho_p r^2 u_0/9\mu R \quad \text{(Stokes number)}$$

$$U_x = u_x/u_0$$

$$U_y = u_y/u_0$$

$$A_x = \frac{F_{D-x}}{m}\frac{R}{u_0}$$

$$A_y = \frac{F_{D-y}}{m}\frac{R}{u_0}.$$

where R is the collector (droplet) radius and X, Y, V_x, Y_y, U_x, U_y, T, A_x, A_y are all nondimensional.

The resulting nondimensional equations are

$$\frac{d^2X}{dT^2} = \frac{1}{K}(U_x - V_x) + A_x \tag{30}$$

$$\frac{d^2Y}{dT^2} = \frac{1}{K}(U_y - V_y) + A_y \tag{31}$$

with initial conditions

$$T = 0,\ X = -\infty,\ Y = Y^0.$$

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EFFECT OF DIFFUSIOPHORESIS ON PARTICLE COLLECTION BY WET SCRUBBERS*

THE THEORETICAL calculations of scrubber efficiencies resulting from the combined effects of inertial impaction and diffusion presented by L. E. SPARKS and M. J. PILAT are interesting in that they attempt to determine the influence of condensation on the separation effect on which there was no previous quantitative information. The answer to this problem is very important for the conditions of operation. Certainly the effect of condensation due to local temperature gradients not only promotes particle collection on droplets of lower temperature by means of vapour diffusion but also influences other processes like the formation of water films upon particles acting as condensation cores and so increases particle masses. Therefore, an increase in scrubber efficiencies of separation as a result of condensation can be expected. However, it is surprising that these calculations indicate such large differences of overall efficiencies of collection between the case of no mass transfer and that of condensation within the field of particles of size of about 1 μm (FIG. 2). To my knowledge this appears to be at variance with experience and observation. The influence of condensation should decrease rapidly with increasing particle size as a consequence of the decrease of drag effect by diffusion (A. T. LITWINOW, *J. appl. Chem. U.S.S.R.* **40** (2), 335–342, 1967) so that the values of efficiencies overlap.

The calculations should be supported by some additional observations.

Universität Stuttgart
Stuttgart
Germany

CHRISTIAN ALT

AUTHORS' REPLY

WE AGREE with Professor ALT that the effect of diffusiophoresis on the particle collection efficiency of wet scrubbers decreases rapidly with increasing particle size. In our paper we reported that the calculated difference in the collection efficiency of the 100 μm radius droplet moving through air at 100 cm s^{-1} between condensation and evaporation with a water vapor pressure gradient of 500 mbar cm^{-1} is more than an order of magnitude for a 1 μm radius particle and less than 1 per cent for an 18 μm radius particle.

However, we disagree with Professor ALT's statement that our calculated results appear to be at variance with experience and observations (no references of reported observations were provided). Our calculations appear to be in qualitative agreement with the experimental observations cited in our paper. We pointed out that LAPPLE and KAMACK (1955) reported that the addition of steam (2–3 times that required to saturate the air at room temperature) produced a fivefold reduction in the dust loss in their semiworks pipeline contactor at a given pressure drop.

It should be noted that the calculated results for FIGS. 1, 2, and 3 are for the specific scrubber case indicated in the paper and thus do not apply to every situation.

We would like to take this opportunity to point out a typographical error in the Appendix of the paper. The exponent 2 for u_0 (the undisturbed fluid velocity) was omitted in the expression for the nondimensional acceleration. The equations for A_x and A_y should be

$$A_x = \frac{F_{D-x}}{m}\frac{R}{{u_0}^2}$$

$$A_y = \frac{F_{D-y}}{m}\frac{R}{{u_0}^2}$$

Water and Air Resources Division
Department of Civil Engineering
University of Washington
Seattle, Washington 98105

LESLIE E. SPARKS and
MICHAEL J. PILAT

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