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CONVECTIVE MASS OR HEAT TRANSFER FROM SIZE-DISTRIBUTED DROPS, BUBBLES OR SOLID PARTICLES

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Abstract—Available analyses of convective mass or heat transfer in particulate systems treat only a *single* drop or bubble in the ideal case of a completely pure system. There is, therefore, a definite need to establish correlations for *assemblages* of many drops or bubbles in practical two-phase systems where surfactant impurities are ever present. Such correlations are developed in the present work by employing the von Kármán–Pohlhausen integral method to the boundary layer formed in moving clouds of spherical drops, bubbles, or solid particles. The results obtained generalize previous analyses for single particles and for assemblages of *solid* particles. In particular this method allows the evaluation of the yet untreated practical cases of convective transfer in intermediately circulating ensembles of drops or bubbles. Employing these new results the applicability of the method is extended to the analysis of *total* average convective transfer in size-distributed populations. For the establishment of an appropriate particle size distribution function a large number of experimental data are correlated by a single parameter function which is readily integrated to give total average interfacial transfer rates. A comparison with other correlations demonstrates the generality and applicability of the present results which include to a first approximation most available correlations for drops or bubbles at rapid, intermediate, and zero internal circulation.

NOMENCLATURE

a,	radius of a typical particle in the population;
$\bar{a}_3$,	mean volume radius defined by equation (26);
$\bar{a}_{32}$,	surface mean radius defined by equation (29);
B,	coefficient in equation (35);
c,	concentration in the diffusional boundary layer around drops or bubbles (binary system);
C_δ,	concentration in the bulk fluid and at the edge of boundary layer;
C_i,	equilibrium concentration at interface of particle;
D,	binary diffusion coefficient;
$f(a)$,	a normalized particle size distribution function (number density);
k,	average mass transfer coefficient for uniform sized system;
$\bar{k}_3$,	average mass transfer coefficient (based on $\bar{a}_3$) for size-distributed system;
L,	average linear dimension of impeller or dispersing device;
M,	total average interfacial transfer rate over the entire population of particles defined by equation (24);
N,	total number of particles in the particulate system defined by equation (25);
U,	relative velocity between continuous and dispersed phases (see [25] for definition);
U_e,	ensemble velocity defined by equation (18);
U_0,	interfacial velocity at equator of particle;

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U_s,	Stokes' velocity of single solid particle;
V,	total volume of the particulate system;
V_r,	radial component of velocity vector in continuous phase;
V_θ,	tangential component of velocity vector in continuous phase;
W,	function defined by equation (6);
y,	normal distance to particle surface $y = (r - a)$;
Y,	function defined by equation (7).

Greek symbols

α,	parameter defined by equation (32);
β,	viscosity ratio defined by equation (8);
γ,	"interfacial viscosity" due to adsorbed surfactant impurities;
$\delta(\theta)$,	thickness of diffusional boundary layer;
δ_m,	mean thickness of diffusional boundary layer;
η,	dimensionless radius defined by equation (9);
θ,	cone angle between radius vector and the vertical directed in the sense of the drop motion;
μ,	viscosity;
ρ,	density;
σ,	interfacial tension;
ϕ,	volume fraction occupied by dispersed phase;
Ω,	rotational speed of impeller.

Dimensionless numbers

Pe,	$2Ua/D$;
Pe_e,	$2U_e a/D$;
$\overline{Pe}_3$,	$2U_e \bar{a}_3/D$;
Sh,	$2ak/D$;
$\overline{Sh}_3$,	$2\bar{a}_3\bar{k}_3/D$;
We,	$L^3\rho\Omega^2/\sigma$.

Superscripts

c,	refers to the continuous phase;
d,	refers to the dispersed phase.

INTRODUCTION

THE ESTIMATION of convective interfacial mass or heat transfer rates in assemblages of many drops, bubbles, or solid particles is very complicated since it requires a simultaneous analysis of a large number of independent variables. A considerable contribution to these complications is due to the combined effects of particle concentration, surfactant concentration, and particle size distribution on the transfer mechanisms in such systems. In practical applications (where surfactant impurities are ever present) all three effects play a significant role in changing the velocity, temperature and concentrations fields from those predicted for a single particle in a pure system. Therefore, in analysing the fluid mechanics, heat, and mass transfer mechanisms of these systems the last three effects should be taken into account. Unfortunately most available analyses treat only a single drop, bubble, or solid particle in the ideal case of a completely pure interface (see Table 1).

The aim of this work is to evaluate convective interfacial mass (or heat) transfer rates to or from ensembles of many spherical drops, bubbles, or solid particles (uniform and size distributed) at high Péclét and low particle Reynolds numbers. For that purpose we employ here the thin boundary-layer concept coupled with an integral-method approach. similar to the von Kármán–Pohlhausen method. The results obtained are then compared with available correlations in some limited domains and with the Levich boundary layer approach [11] extended recently by Waslo and Gal-Or [18] for ensembles of drops and bubbles. In the last paper as well as in the present one the velocity fields derived earlier by Gal-Or and Waslo [8] are employed. For the special case of ensembles of solid particles these velocity fields reduce to those of Happel [10]. The latter were employed by Ruckenstein [14] and Pfeffer [13] to evaluate convective transfer in ensembles of (uniform) spherical solid particles (where surfactant impurities play no role and internal circulation is absent). It would, therefore, be of particular interest to compare the results of the present work for ensembles of drops or bubbles with those of Ruckenstein and Pfeffer for ensembles of solid particles.

Table 1. Comparison of available correlations with present results for convective mass transfer rates to or from single or assemblāges of drops, bubbles, or solid particles at low particle Reynolds numbers

Expression	Particles	Internal circulation	Pe	Reference
$Sh = 0{\cdot}923\left(\beta\dfrac{1-\phi^{\frac{5}{3}}}{Y-\phi^{\frac{1}{3}}W}\right)^{\frac{1}{2}}Pe_e^{\frac{1}{2}}$	Ensemble	Strong	Large	Waslo and Gal-Or [18]
$Sh = 0{\cdot}895\left(\beta\dfrac{1-\phi^{\frac{5}{3}}}{Y-\phi^{\frac{1}{3}}W}\right)^{\frac{1}{2}}Pe_e^{\frac{1}{2}}$	Ensemble	Strong	Large	Present work
$0{\cdot}849\left(\beta\dfrac{1-\phi^{\frac{5}{3}}}{Y}\right)Sh^{-2} + 4{\cdot}96\left\{\dfrac{2}{15}\left[\dfrac{3}{2} - 2\beta\dfrac{(1-\phi^{\frac{5}{3}})}{Y}\right] + \phi^{\frac{5}{3}}\left[\beta\dfrac{(1-\phi^{\frac{5}{3}})}{Y} - \dfrac{1}{2}\right]\right\}Sh^{-3} = Pe^{-1}$	Ensemble	Intermediate	Large	Present work
$Sh = 1{\cdot}26\left(\dfrac{1-\phi^{\frac{5}{3}}}{Y-\phi^{\frac{1}{3}}W}\right)^{\frac{1}{3}}Pe_e^{\frac{1}{3}}$	Ensemble	None	Large	Pfeffer [13]
$Sh = 1{\cdot}306\left(\dfrac{1-\phi^{\frac{5}{3}}}{Y-\phi^{\frac{1}{3}}W}\right)^{\frac{1}{3}}Pe_e^{\frac{1}{3}}$	Ensemble	None	Large	Present work
$Sh = 1{\cdot}5\left(\dfrac{1-\phi^{\frac{5}{3}}}{Y-\phi^{\frac{1}{3}}W}\right)^{\frac{1}{3}}Pe^{\frac{1}{3}}$	Ensemble	None	Large	Ruckenstein [14]
$Sh = 0{\cdot}923\left(\dfrac{U_0}{U_s}\right)^{\frac{1}{2}}Pe^{\frac{1}{2}}$	Single	Strong	Large	Levich [11]
$Sh = 0{\cdot}923\left(\dfrac{U_0}{U_s}\right)^{\frac{1}{2}}Pe^{\frac{1}{2}}$	Single	Strong	Large	Lochiel and Calderbank [12]
$Sh = 0{\cdot}895\left(\dfrac{U_0}{U_s}\right)^{\frac{1}{2}}Pe^{\frac{1}{2}}$	Single	Strong	Large	Ruckenstein [14]
$Sh = 0{\cdot}948\left(\dfrac{U_0}{U_s}\right)^{\frac{1}{2}}Pe^{\frac{1}{2}}$	Single	Strong	Large	Griffith [9]
$Sh = 0{\cdot}864\left(\dfrac{U_0}{U_s}\right)^{\frac{1}{2}}Pe^{\frac{1}{2}}$	Single	Strong	Large	Ward, Trass and Johnson [17]
$0{\cdot}849\left(\dfrac{U_0}{U_s}\right)Sh^{-2} + 0{\cdot}662\left(\dfrac{3}{2} - 2\dfrac{U_0}{U_s}\right)Sh^{-3} = Pe^{-1}$	Single	Intermediate	Large	Ruckenstein [14]
$Sh = 0{\cdot}998\,Pe^{\frac{1}{3}}$	Single	None	Large	Levich [11]
$Sh = 0{\cdot}99\,Pe^{\frac{1}{3}}$	Single	None	Large	Lochiel and Calderbank [12]
$Sh = 0{\cdot}99\,Pe^{\frac{1}{3}}$	Single	None	Large	Friedlander [4]
$Sh = 1{\cdot}037\,Pe^{\frac{1}{3}}$	Single	None	Large	Akselrud [1]
$Sh = 0{\cdot}89\,Pe^{\frac{1}{3}}$	Single	None	Large	Friedlander [21]
$Sh = 1{\cdot}037\,Pe^{\frac{1}{3}}$	Single	None	Large	Ruckenstein [14]
$Sh = 2\left(1 + \dfrac{Pe}{4} + 0{\cdot}15\,Pe^2 + \ldots\right)$	Single	None	Small	Kronig and Bruijsten [22]
$Sh = 2(1 + 0{\cdot}16\,Pe^2)$	Single	None	Small	Frisch [23]

Table 1—*continued*

Expression	Particles	Internal circulation	Pe	Reference
$Sh = 2\left(1 + \frac{Pe}{4} + \frac{Pe^2}{12} + \ldots\right)$	Single	None	Small	Friedlander [21]
M, Total average interfacial transfer given by equation (37)	Size—distributed ensemble	Strong	Large	Present work
M, Total average interfacial transfer given by equation (38)	Size—distributed ensemble	None	Large	Present work

CONVECTIVE TRANSFER IN ENSEMBLES OF UNIFORM PARTICLES

Mass and heat transfer between moving drops, bubbles, or solid particles is an inherently unsteady-state process. However, small particles moving at low particle Reynolds numbers attain their terminal velocity in a few particle diameters after detachment from their formation devices. If changes of particle mass due to diffusion and possible changes in surface tension, viscosities, etc., are negligible, steady-state analysis becomes valid for times greater than that required for the particle to attain its steady-state velocity. The mass transfer resistance inside drops or bubbles can be neglected when its contents consists of only one chemical component (and the continuous phase is insoluble in the dispersed phase), or when the combination of diffusivity and internal circulation makes the internal resistance negligible compared to that of the continuous phase. Levich [4] demonstrated that even at $Re \ll 1$ a thin diffusional boundary layer is formed around the droplet interface provided that $Pe \gg 1$—a condition preveailing in most liquid phases. Under these conditions and when axisymmetrical flow exists around the particles of the assemblage [8] the concentration fields in the binary diffusional boundary layer are governed by*

$$V_r\frac{\partial c}{\partial y} + \frac{V_\theta}{a}\frac{\partial c}{\partial \theta} = D\frac{\partial^2 c}{\partial y^2}, \quad y = r - a; y \ll 1 \quad (1)$$

subject to the boundary conditions

$$\begin{aligned} c &= C_i \quad \text{for} \quad y = 0 \\ c &= C_\delta \quad \text{for} \quad y = \delta(\theta) \end{aligned} \quad (2)$$

where $\delta(\theta)$ is the thickness of the diffusional (or thermal) boundary layer.

Integration of equation (1) with respect to y between the limits 0 and $\delta(\theta)$ combined with the continuity equation can readily lead to

$$\begin{aligned} -D\left(\frac{\partial c}{\partial y}\right)_{y=0} &= \frac{1}{a}\frac{\partial}{\partial \theta}\int_0^{\delta(\theta)} (c - C_\delta)\, V_\theta\, \mathrm{d}y \\ &+ \frac{1}{a}\cot\theta \int_0^{\delta(\theta)} (c - C_\delta)\, V_\theta\, \mathrm{d}y. \end{aligned} \quad (3)$$

where the term $2V_r/r$ in the continuity equation has been neglected.

We now approximate the concentration field in the boundary layer by the polynomial

$$\frac{c - C_\delta}{C_i - C_\delta} = 1 - 2\frac{y}{\delta(\theta)} + 2\left(\frac{y}{\delta(\theta)}\right)^3 - \left(\frac{y}{\delta(\theta)}\right)^4. \quad (4)$$

For the velocity component V_θ in the phase surrounding ensembles of drops, bubbles or solid particles we employ the Gal-Or and Waslo [8] velocity field

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* The analysis and formulations for convective heat transfer is similar and, therefore, will not be repeated here.

$$V_\theta = \frac{3}{2}\frac{U_s}{W}\left(-\frac{1}{2\eta^3} - \frac{1}{2\eta}W + Y - 2\phi^{1/3}\eta^2\right)\sin\theta, \tag{5}$$

where

$$W = 3 + 2\beta + 2\phi^{1/3}(1 - \beta), \tag{6}$$

$$Y = 2 + 2\beta + \phi^{1/3}(3 - 2\beta), \tag{7}$$

$$\beta = \frac{\mu^c}{\mu^d + \gamma}, \tag{8}$$

$$\eta = 1 + \frac{y}{a}. \tag{9}$$

Here ϕ is the volume fraction occupied by the dispersed phase and γ the "interfacial viscosity" due to surfactant impurities. The last is responsible for retardation of internal circulation in drops or bubbles. The derivation and physical significance of γ has been fully discussed elsewhere [8].

Rewriting V_θ in terms of $U_0 \equiv V_\theta$ ($y = 0$, $\theta = \pi/2$, any ϕ) and $U_s \equiv V_\theta$ ($y \to \infty$, $\theta = \pi/2$, $\phi \to 0$) one easily obtains

$$V_\theta = \left[\frac{(U_0 - U_s/2)}{\phi^{1/3} + 2}\left(1 + \frac{y}{a}\right)^{-3} + \frac{(\frac{3}{2}U_s - U_0 + U_0\phi^{1/3})}{\phi^{1/3} + 2}\left(1 + \frac{y}{a}\right)^{-1} + U - 2\phi^{1/3}\frac{(U_s - 2U_0)}{\phi^{1/3} + 2}\left(1 + \frac{y}{a}\right)^2\right]\sin\theta \tag{10}$$

Neglecting terms containing higher powers than one of y/a and $\phi^{1/3}$, as well as their products, equation (10) reduces to

$$V_\theta = \left[U_0 + \frac{y}{a}(\tfrac{3}{2}U_s - 2U_0) + \frac{15}{2}\frac{y}{a}\phi^{1/3}(U_0 - \tfrac{1}{2}U_s)\right]\sin\theta. \tag{11}$$

Bringing equations (4) and (11) to (3) one can show that

$$\frac{\mathrm{d}[\delta(\theta)/a]}{\mathrm{d}\theta} = \frac{\dfrac{4}{Pe\sin\theta}\dfrac{a}{\delta(\theta)} - \left[\dfrac{3}{5}\dfrac{\delta(\theta)}{a}\dfrac{U_0}{U_s} + \dfrac{2}{15}\left(\dfrac{\delta(\theta)}{a}\right)^2\left(\dfrac{3}{2} - 2\dfrac{U_0}{U_s}\right) + \phi^{1/3}\left(\dfrac{\delta(\theta)}{a}\right)^2\left(\dfrac{U_0}{U_s} - \dfrac{1}{2}\right)\right]\cot}{\dfrac{3}{10}\dfrac{U_0}{U_s} + \dfrac{2}{15}\dfrac{\delta(\theta)}{a}\left(\dfrac{3}{2} - 2\dfrac{U_0}{U_s}\right) + \phi^{1/3}\dfrac{\delta(\theta)}{a}\left(\dfrac{U_0}{U_s} - \dfrac{1}{2}\right)} \tag{12}$$

Defining the average value of convective mass transfer coefficient over the whole surface of a particle by

$$k = \frac{-D\int_0^\pi [(\partial c/\partial y)]_{y=0}\, 2\pi a^2 \sin\theta\,\mathrm{d}\theta}{4\pi a^2(C_i - C_\delta)} \tag{13}$$

and using equation (4) together with the definition $Sh = 2ak/D$, one obtains

$$Sh = 2a\int_0^\pi \frac{\sin\theta}{\delta(\theta)}\mathrm{d}\theta. \tag{14}$$

For the following analysis we now distinguish between three main domains of internal circulation:

(i) Rapidly circulating drops or bubbles (i.e. in highly purified flows in the laboratory).
(ii) Non-circulating solid particles or drops and bubbles (the last two systems are known by observations to behave like solid particles even when the contamination degree is relatively low [20, 24]).
(iii) Intermediate rates of internal circulation (covers most practical cases where internal circulation in drops and bubbles is only partially retarded).

(i) *Rapidly circulating drops or bubbles*

For rapid circulating drops or bubbles, namely when

$$\frac{3}{10}\frac{U_0}{U_s} \gg \frac{2}{15}\frac{\delta_m}{a}\left(\frac{3}{2} - \frac{U_0}{U_s}\right) + \phi^{\frac{5}{3}}\frac{\delta_m}{a}\left(\frac{U_0}{U_s} - \frac{1}{2}\right),$$

equation (12) can be approximated by

$$\frac{\delta(\theta)}{a}\cdot\frac{\mathrm{d}[\delta(\theta)/a]}{\mathrm{d}\theta} = \frac{40}{3\,Pe\sin\theta}\frac{U_s}{U_0} - 2\left(\frac{\delta(\theta)}{a}\right)^2\cot\theta. \qquad (15)$$

Integrating equation (15) and applying the condition that $\delta(\theta)$ approaches a finite value when $\theta \to 0$, gives

$$\frac{\delta(\theta)}{a} = \left(\frac{80}{9}\right)^{1/2} Pe^{-1/2}\frac{(\cos^3\theta - 3\cos\theta + 2)^{1/2}}{\sin^2\theta}\left(\frac{U_0}{U_s}\right)^{-1/2}. \qquad (16)$$

Introducing this solution into equation (14) and integrating finally gives

$$Sh = 0{\cdot}895\left(\frac{U_0}{U_s}\right)^{1/2} Pe^{1/2}$$

$$= 0{\cdot}895\left(\beta\frac{1 - \phi^{\frac{5}{3}}}{Y}\right)^{1/2} Pe^{1/2}. \qquad (17)$$

Waslo and Gal-Or [18] defined Péclét number on a basis of "ensemble velocity"

$$U_e = -\tfrac{3}{2}U_s\left(\phi^{\frac{1}{3}} - \frac{Y}{W}\right). \qquad (18)$$

Using their definition, equation (17) becomes

$$Sh = 0{\cdot}895\left(\beta\,\frac{1 - \phi^{\frac{5}{3}}}{Y - \phi^{\frac{1}{3}}W}\right)^{1/2} Pe_e^{1/2}, \qquad (19)$$

which is similar in form to that derived by Waslo and Gal-Or by a more exact analysis (see Table 1). When $\phi \to 0$ and $\gamma \to 0$, equation (19) reduces to

$$Sh = 0{\cdot}63\left(\frac{\mu^c}{\mu^d + \mu^c}\right)^{1/2} Pe^{1/2} \qquad (20)$$

which is identical with the expressions of Ruckenstein [14], for convective mass transfer to a single clean drop or bubble. Levich [11] as well as Lochiel and Calderbank [12], found by using a more exact method, the numerical coefficient in equation (20) to equal 0·65, while Griffith [9], and Ward *et al.* [17], who assumed different concentration profiles, give the values 0·67 and 0·61 respectively.

(ii) *Solid particles and non-circulating drops or bubbles*

For negligible values of U_0/U_s namely, when

$$\frac{U_0}{a}\left(\frac{3}{5} - \frac{4}{15}\frac{\delta_m}{a} + \phi^{\frac{5}{3}}\frac{\delta_m}{a}\right) \ll \frac{\delta_m}{a}\left(\frac{1}{5} - \frac{1}{2}\phi^{\frac{5}{3}}\right),$$

equation (12) can be approximated by

$$\frac{\mathrm{d}[\delta(\theta)/a]}{\mathrm{d}\theta} = \frac{20}{Pe\sin\theta}\left(\frac{a}{\delta(\theta)}\right)^2(1 - \tfrac{5}{2}\phi^{\frac{5}{3}})^{-1} - \frac{\delta(\theta)}{a}\cot\theta.$$

Integrating this equation and again applying the condition that $\delta(\theta)$ approaches a finite value when $\theta \to 0$, one obtains

$$\frac{\delta(\theta)}{a} = (15)^{1/3}(1 - \tfrac{5}{2}\phi^{\frac{5}{3}})^{-1/3} Pe^{-1/2}\frac{(2\theta - \sin 2\theta)^{1/3}}{\sin\theta}$$

Introducing this solution into equation (14) and using the approximation

$$1 - \tfrac{5}{2}\phi^{\frac{5}{3}} \approx \frac{1 - \phi^{\frac{5}{3}}}{1 + \frac{3}{2}\phi^{\frac{5}{3}}},$$

one obtains in terms of U_e

$$Sh = 1{\cdot}306\left(\frac{1 - \phi^{\frac{5}{3}}}{Y - \phi^{\frac{1}{3}}W}\right)^{\frac{1}{3}} Pe_e^{\frac{1}{3}}, \qquad (21)$$

and in terms of U

$$Sh = 1{\cdot}037\left(\frac{1 - \phi^{\frac{5}{3}}}{1 + \frac{3}{2}\phi^{\frac{5}{3}}}\right)^{\frac{1}{3}} Pe^{\frac{1}{3}}. \qquad (22)$$

Equation (21) is similar in form to that, derived by Waslo and Gal-Or [18], and Pfeffer [13], who give the value of 1·26 for the numerical coefficient. Equation (22) is also similar to that obtained by Ruckenstein [14] for ensembles of rigid spheres. When $\phi \rightarrow 0$, equation (22) reduces to

$$Sh = 1{\cdot}037\, Pe^{\frac{1}{3}}$$

which is identical with the expression of Ruckenstein [14] and Akselrud [1] for single solid spheres, and is similar in form to that of Levich [11], Friedlander [4], and Lochiel and Calderbank [12], whose numerical coefficient is 0·998, (see Table 1).

Our results show that the Sherwood number in particle ensembles decreases with increasing volume concentrations of the dispersed phase, as well as with increasing values of γ (due to retardation of internal circulation by impurities). The last effect is a function of β [equation (8)]. The significance of the latter conclusion is apparently not restricted to equation (19), which is valid at high β values.

(iii) *Intermediate rates of internal circulation*

In most practical cases the internal circulation in each member of the drop or bubble ensembles is only partially retarded due to traces of surfactant impurities which are ever present in particle technology (even the equipment or the atmosphere may supply enough impurities to cause a significant change in rates of heat and mass transfer in the system). For the range of intermediate rates of internal circulation (i.e. intermediate values of β or U_0/U_s) one must integrate equation (12) numerically, or else resort to approximate interpolation methods. Such an interpolation has been done in this work whereby equation (12) can be reexpressed as

$$0{\cdot}849\,\frac{U_0}{U_s}Sh^{-2} + 4{\cdot}96\left[\frac{2}{15}\left(\frac{3}{2} - 2\,\frac{U_0}{U_s}\right) + \phi^{\frac{5}{3}}\left(\frac{U_0}{U_s} - \frac{1}{2}\right)\right]Sh^{-3} = Pe^{-1}, \qquad (23a)$$

or in terms of the more practical variables

$$0{\cdot}849\left(\beta\frac{1-\phi^{\frac{5}{3}}}{Y}\right)Sh^{-2} + 4{\cdot}960\left\{\frac{2}{15}\left[\frac{3}{2} - 2\beta\frac{(1-\phi^{\frac{5}{3}})}{Y}\right] + \phi^{\frac{5}{3}}\left[\beta\frac{(1-\phi^{\frac{5}{3}})}{Y} - \frac{1}{2}\right]\right\} Sh^{-3} = Pe^{-1}. \qquad (23b)$$

It is interesting to note that for solid particles or non-circulating drops and bubbles (where $\beta = 0$ or $U_0 = 0$) most correlations of Table 1 agree in showing that

$$Sh \propto Pe^{\frac{1}{3}},$$

while for rapidly circulating drops or bubbles in ideal conditions

$$Sh \propto Pe^{\frac{1}{2}}.$$

These well-known limits as well as the entire practical range of intermediate β values are covered in equation (23).

Furthermore equation (23) generalizes available correlations for a single solid particle, drop or bubble to ensembles which consist of many particles. Hence equation (23) is the most general one available at present for correlating convective mass (or heat) transfer in ensembles of particles in the presence of surfactant impurities. It contains as a special case the expression derived by Ruckenstein [14] for a single particle (see Table 1).

TOTAL AVERAGE CONVECTIVE TRANSFER IN SIZE-DISTRIBUTED PARTICULATE SYSTEMS

A general analysis and formulation for the evaluation of total average coupled heat and multicomponent mass transfer in size-distributed particulate systems has been recently given by Gal-Or [7]. According to this formulation the total average interfacial mass transfer in uncoupled binary systems with large population of particles characterized by any general normalized particle size distribution

$$\int_0^\infty f(a)\,\mathrm{d}a = 1$$

is given by

$$M = -\int_0^{\pi}\int_0^{\infty} ND\left(\frac{\partial c}{\partial y}\right)_{y=0} 2\pi a^2 \sin\theta f(a)\,\mathrm{d}a\,\mathrm{d}\theta, \quad (24)$$

where

$$N = \frac{3\phi V}{4\pi(\bar{a}_3)^3}, \quad (25)$$

$$\bar{a}_3 = \left[\int_0^{\infty} a^3 f(a)\,\mathrm{d}a\right]^{\frac{1}{3}}. \quad (26)$$

Employing equation (4) and (14) in (24) and defining a mean volume Sherwood number,

$$\overline{Sh_3} = \frac{2\,\bar{a}_3\bar{k}_3}{D} \quad (27)$$

one obtains the total average *convective* interfacial transfer rate

$$M = \frac{3}{2}\frac{\phi V}{(\bar{a}_3)^3}\overline{Sh_3}\,(C_i - C_\delta)\int_0^{\infty} af(a)\,\mathrm{d}a. \quad (28)$$

Here Sherwood number has not been included in the integral. This is based on previous conclusion of Gal-Or and Hoelscher [5] which states that negligible error is introduced in the mass- (or heat)-transfer coefficient if one replaces the variable particle size by the appropriate statistical mean.

To integrate equation (28) one must employ the appropriate expression for $f(a)$. For many dispersed systems such expressions are rather complicated and do not readily lend themselves to analytical treatment. In certain cases, however, such as that of some gas–liquid and liquid–liquid dispersions produced by mechanical agitation, one can approximate the size-distribution function by simpler relations, containing only a single parameter.

Such a practical case is illustrated in Fig. 1, where some experimental data are correlated by using a dimensionless radius $a/\bar{a}_{32}$, where $\bar{a}_{32}$ is the surface mean radius defined by

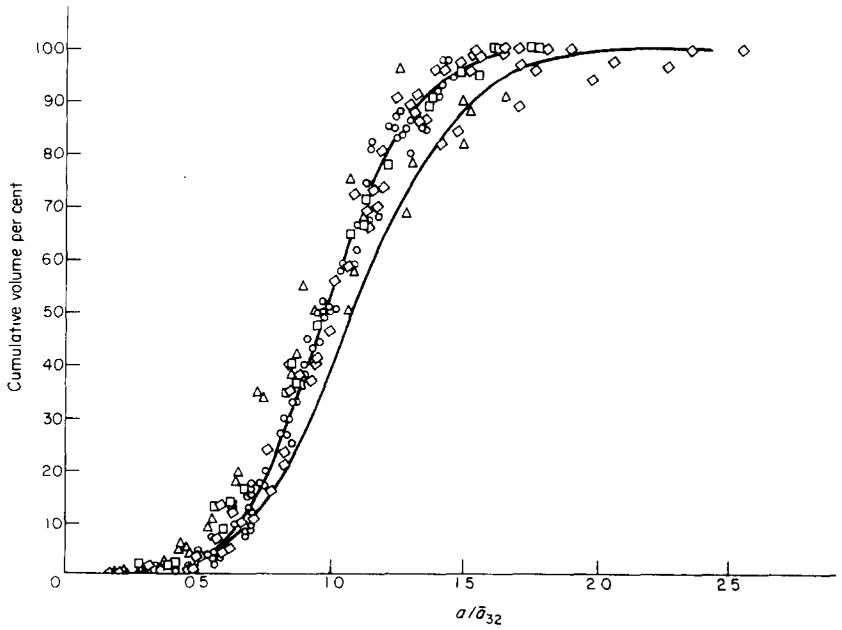

FIG. 1. Generalized size distribution data for gas–liquid and liquid–liquid dispersions.
○ Experimental data of Chen and Middleman [3] for liquid–liquid dispersions in agitated Vessels;
△ Data of Gal-Or [6] for gas–liquid dispersions in agitated contractor;
□ Data of Weaver *et al.* [19] for isobutanol-in-water dispersions in a spray column; and
· Data of Shinnar [16] for molten wax–water dispersions in stirred vessels.
——— Bayens distribution function [equation (31)].
– – – – Chen and Middleman's empirical expression [equation (30)].

$$\bar{a}_{32} = \frac{\int\limits_0^\infty a^3 f(a)\,\mathrm{d}a}{\int\limits_0^\infty a^2 f(a)\,\mathrm{d}a}. \qquad (29)$$

The data include those of Chen and Middleman [3] for dispersions of 14 different organic liquids in water in agitated vessels of different diameters and agitator-to-vessel dameter ratios; of Gal-Or [6, 7] for air–water dispersions in agitated contactors at different agitator speeds; of Shinnar [16] for dispersions of 5% molten Shellwax in hot water in the presence of 0·1% polyvinyl alcohol, produced in an agitated vessel by a turbine mixer rotating at different speeds; and the data of Weaver *et al.* [19] for dispersion of isobutanol in water, obtained by passing the organic phase through $\frac{1}{8}$ in. nozzles into a spray column.

As seen from Fig. 1, the data are adequately correlated by the empirical expression of Chen and Middleman [3]

$$f(a) = \frac{1}{0{\cdot}23\sqrt{\pi}} \exp\left[-9{\cdot}2\left(\frac{a}{\bar{a}_{32}} - 1{\cdot}06\right)^2\right]. \qquad (30)$$

To evaluate total interfacial mass transfer rate from equation (28) one would, however, prefer to approximate the size-distribution data by the more readily integrable expression

$$f(a) = 4\left(\frac{\alpha^3}{\pi}\right)^{\frac{1}{2}} a^2 \exp\left[-\alpha a^2\right], \qquad (31)$$

originally proposed by Bayens [2] for coagulation of hydrosols in Brownian motion, and subsequently modified by Gal-Or and Hoelscher [5]. In equation (31),

$$\alpha = \left(\frac{4}{(\sqrt{\pi})(\bar{a}_3)^3}\right)^{\frac{2}{3}}. \qquad (32)$$

It has been shown [7], that for this case

$$\bar{a}_3 = \bar{a}_{32}/1{\cdot}148. \qquad (33)$$

Equation (31) has the same form as the Maxwell–Boltzmann speed distribution function for gaseous atoms, and may conceivably be associated with the spectrum of energy levels involved in the process of dispergation.

Introducing equation (31) into (28) and integrating one finally obtains the general design equation

$$M = \frac{2{\cdot}2}{(\bar{a}_3)^2}\,\overline{Sh_3}\,\phi V(C_\delta - C_i). \qquad (34)$$

Shinnar and Church [15] have shown theoretically that for dispergation by mechanical agitation, when break-up of droplets is the predominant mechanism with respect to their coalescence, the surface mean radius may be related to the agitator Weber number by

$$\bar{a}_{32} = BL(We)^{-0{\cdot}6} \qquad (We \equiv L^3\rho\Omega^2/\sigma). \quad (35)$$

Their prediction was experimentally substantiated by Chen and Middleman [3], who found $B = 0{\cdot}026$ for small values of ϕ.

Employing equation (33) and (35) and the above value of B in (34) one obtains the practical result

$$M = 4{\cdot}25 \times 10^3 \,.\, \frac{(We)^{1{\cdot}2}}{L^2}\,\overline{Sh_3}\,\phi V(C_\delta - C_i). \quad (36)$$

For mass transfer from ensembles of drops or bubbles with high internal circulation (highly pure systems), equation (36) becomes

$$M = 3{\cdot}8 \times 10^3 \left(\beta\frac{1 - \phi^{\frac{5}{3}}}{Y - \phi^{\frac{1}{3}}W}\right)^{\frac{1}{2}} \times \frac{(\overline{Pe}_3)^{\frac{1}{2}}(We)^{1{\cdot}2}}{L^2}\,\phi V(C_\delta - C_i), \quad (37)$$

and for ensembles of solid particles or non-circulating drops or bubbles

$$M = 5{\cdot}55 \times 10^3 \left(\frac{1 - \phi^{\frac{5}{3}}}{Y - \phi^{\frac{1}{3}}W}\right)^{\frac{1}{3}} \frac{(\overline{Pe}_3)^{\frac{1}{3}}(We)^{1{\cdot}2}}{L^2}\,\phi V(C_\delta - C_i). \quad (38)$$

For intermediately circulating size-distributed drops or bubbles one must evaluate $\overline{Sh_3}$ from equation (23). This can be done when the particular operating conditions are known (i.e. ϕ, ρ_c, ρ_d, μ_c, μ_d, γ, $\bar{a}_3$ and g). These equations can be used in predicting trends in operating conditions and efficiency of size-distributed two-phase dispersed systems.

Table 1 compares the present results with available correlations for convective transfer rates to or from single drops, bubbles or solid particles or their assemblages moving at low particle Reynolds numbers. The comparison demonstrates the generality and applicability of the present results which include to a first approximation most available correlations for single particles and assemblages moving with rapid, intermediate and zero internal circulation.

Finally it should be noted that the Sherwood numbers reported here do not include the non-flow contribution which can usually be approximated by $2/(1 - \phi^{\frac{1}{3}})$. A more exact evaluation of the non-flow contribution is available elsewhere [26].

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CONVECTION THERMIQUE OU MASSIQUE AUTOUR DE GOUTTES, DE BULLES OU PARTICULES SOLIDES DISTRIBUÉES EN TAILLE

Résumé—Des analyses utilisables de la convection thermique ou massique dans des systèmes à particules traitent seulement d'une goutte ou d'une bulle unique dans le cas idéal d'un système complètement pur. Il est cependant nécessaire d'établir des relations pour des ensembles de plusieurs gouttes ou bulles dans des systèmes pratiques à deux phases où les impuretés surfactantes sont toujours présentes. De telles formules sont développées dans cet article en appliquant la méthode intégrale de Von Kármán–Pohlhausen à une couche limite formée dans des nuages mobiles de gouttes, bulles ou particules solides sphériques. Les résultats obtenus généralisent des analyses antérieures pour des particules uniques et pour des ensembles de particules solides. En particulier, cette méthode permet l'évaluation de cas pratiques, encore non traités, de la convection dans des ensembles de gouttes ou bulles circulant en régime intermédiaire. A partir de ces nouveaux résultats, l'application de cette méthode est étendue à l'analyse de la convection globale moyenne dans des populations distribuées en taille. Pour l'établissement d'une fonction appropriée de distribution en taille des particules, un grand nombre de résultats expérimentaux est unifié par une fonction à paramètre unique qui est déjà intégrée pour donner les flux globaux de transfert interfacial. Une comparaison avec d'autres formules démontre la généralité et l'applicabilité des résultats présents qui incluent en première approximation des formules plus utilisables pour des gouttes ou bulles dans le cas d'une circulation interne rapide, intermédiaire ou nulle.

STOFF- ODER WÄRMEÜBERTRAGUNG VON GRÖSSENVERTEILTEN TROPFEN BLASEN ODER FESTEN TEILCHEN BEI KONVEKTION

Zusammenfassung—Verfügbare Untersuchungen der konvektiven Stoff- oder Wärmeübertragung in Systemen mit einzelnen Teilchen behandeln nur einen einzigen Tropfen oder eine einzige Blase für den Idealfall eines vollkommen reinen Systems. Daher ergibt sich eine gewisse Notwendigkeit, Beziehungen für Anhäufungen vieler Tropfen oder Blasen für in der Praxis vorkommende zweiphasige Systeme aufzustellen, wo Unreinheiten stets vorhanden sind. Solche Beziehungen werden in der vorliegenden Arbeit unter Anwendung des von-Kármán–Pohlhausen-Integrals auf die Grenzschicht entwickelt, wie sie sich in einer bewegenden Menge kugelförmiger Tropfen, Blasen oder fester Teilchen ausgebildet hat. Die erzielten Ergebnisse gestatten eine Verallgemeinerung der vorhergehenden Lösung für einzelne Tropfen und für eine Anhäufung fester Teilchen. Insbesondere erlaubt diese Methode die Auswertung noch unbehandelter praktischer Fälle der konvektiven Übertragung bei untereinander zirkulierenden Tropfen- oder Blasenhaufen. Mit diesen neuen Ergebnissen wird die Anwendbarkeit dieser Methode auf die Untersuchung der gesamten durchschnittlichen konvektiven Übertragung bei grössenverteilter Anhäufung erweitert. Für die Aufstellung einer passenden Verteilungsfunktion für die Teilchengrösse wird eine grosse Anzahl von Versuchswerten herangezogen.

КОНВЕКТИВНЫЙ МАССО- ИЛИ ТЕПЛООБМЕН КАПЕЛЬ, ПУЗЫРЕЙ ИЛИ ТВЕРДЫХ ЧАСТИЦ, РАСПРЕДЕЛЕННЫХ ПО РАЗМЕРУ

Аннотация—Существующий метод анализа конвективного массоили теплообмена в системах, состоящих из частиц, состоит из рассмотрения только одной капли или пузырька для идеального случая чистой системы. Следовательно, существует необходимость установления соотношений для «совокупностей» множества капель или пузырьков в 2-х фазных системах, где постоянно присутствуют сопутствующие загрязнения. Эти соотношения разрабатываются в настоящей работе при использовании интегрального метода Кармана-Польхаузена для пограничного слоя, образующегося в движущихся облаках сферических капель, пузырьков или твердых частиц. Полученные результаты являются обобщением предшествующих анализов для одиночных частиц и совокупностей, состоящих из твердых частиц. В частности, этот метод позволяет оценить до сих пор неисследованные практические случаи конвективного обмена в циркулирующих группах капель или пузырьков. Используя эти новые данные, этот метод мохно применить для анализа суммарного среднего конвективного обмена в популяциях, распределенных по размеру.

Для опредления соответсвующей функции распределения по размеру частиц обобщается большое количество экспериментальных ланных.