

COUPLED HEAT AND MULTICOMPONENT MASS TRANSFER IN PARTICULATE SYSTEMS WITH RESIDENCE TIME AND SIZE DISTRIBUTIONS

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(Received 16 May 1967 and in revised form 7 August 1967)

Abstract—A theory is presented for the general case of coupled heat and multicomponent mass transfer in dispersions and suspensions with residence time and particle size distributions. The general formulations describe the behavior of industrial or biological particulate systems in terms of equations familiar from non-equilibrium thermodynamics and transport phenomena. Using matrix notation and appropriate transformations the resulting partial differential equations are first decoupled and then transformed into ordinary differential equations by employing an integral operator whose kernel takes into account the particle size and residence time distributions. Moment equations of the size distribution are formulated and used to evaluate general interrelationships among average sizes, interfacial surface area, dispersed-phase holdup fraction, and total average rates of transfer from the particle population. The total average heat- and mass-transfer rates in multiphase particulate systems are evaluated in terms of a general normalized size distribution and the expected values of the various fluxes involved. The properties of a proposed general normalized size distribution are discussed and compared with experimental results of bubble size distributions in a gas-liquid dispersion.

NOMENCLATURE

a , radius of a bubble, drop, or solid particle in a population with size distribution;
 \bar{a}_2 , mean surface radius defined by equation (30);
 \bar{a}_3 , mean volume radius defined by equation (28);
 \bar{a}_{32} , surface mean radius defined by equation (27);
 \bar{a}_{43} , volume mean radius defined by equation (29);
 A , total interfacial area of the particle population;
 B , nucleation function which includes rate of formation of new particles by nucleation, agglomeration, breakage, feed and take-off mechanisms;
 B_1, B_2, B_3, B_4 , constants;

B^* , a variable defined by equation (56);
 C_j , mass fraction concentrations, $= \rho_j/\rho$;
 C_p , heat capacity;
 C_{j0} , initial concentration of component j ;
 $f(a)$, a normalized particle size distribution function;
 $f(x_i, r_j, t)$, number density of particles at time t and phase coordinates x_i, r_j ;
 $f(\tau, t)$, age distribution function;
 G_j , growth rate function which depends on concentrations, temperature and internal phase coordinates;
 $I_{\text{ensemble}}^\alpha$, a general integral operator whose kernel takes into account particle size and residence time distributions of the ensemble of particles. It yields total average interfacial transfer rates when operated on local instantaneous gradients at the interface;

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η ,	a measure of the efficiency of mixing defined by equation (42);
μ_i ,	chemical potential;
μ_n ,	the n th moment defined by equation (23);
μ_n^a ,	the n th moment equation when the integration extends from zero to a ;
σ ,	defined by equation (55);
λ ,	defined by equation (54);
λ_b ,	eigenvalues of equation (12);
Λ ,	a diagonal matrix defined by equation (11);
Ψ_{ij} ,	transfer coefficients defined by equations (2) and (3). These coefficients and coupling cross coefficients among the various fluxes involved;
Ω ,	matrix of the transfer coefficients defined by equation (7);
$\bar{\rho}$,	a state vector defined by equation (9);
$\langle \bar{\zeta}(\bar{\tau}) \rangle$,	time average value of ζ over $K(\bar{\tau}, t)$;
ζ_0 ,	initial conditions of ζ ;
ρ_j ,	mass of component j in unit volume of solution;
ρ ,	$= \sum_{j=1}^n \rho_j$ total density;
ω ,	number of times that the time invariant inventory of N particles in the vessel are replaced per unit time.
Superscript	
α ,	denotes the homogeneous phase considered ($\alpha = 1, 2, \dots, \pi$).
Subscripts	
i, j ,	refer to component i or j in an n -component system or to phase space coordinates;
D ,	refers to diffusional flux;
H ,	refers to heat flux.

INTRODUCTION

DISPERSIONS and suspensions are of common

occurrence in many industrial and biological systems and processes such as colloids, blood, aerosols, mist, cloud droplets, biological cells, fermentation, spray drying, spray combustion, emulsion or suspension polymerization, liquid-liquid extraction, direct-contact heat transfer, distillation, boiling, gas absorption, and solid or liquid fluidized beds. In recent years an increasing amount of basic research has been directed to cases involving swarms of bubbles, drops or solid particles which are entrained in a continuous moving phase.

An understanding of the mechanisms of transport processes is essential for the rational design and operation of these particulate systems. This may be gained in part by constructing models amenable to a mathematical treatment. Generally, such a mathematical characterization of a particulate system should include such environmental parameters as composition, temperature and velocity in addition to dispersed-phase holdup volume, particle size and residence time distributions which describe the dependence of particle nucleation and growth on the particle environmental parameters. Subject to the Curie principle [1], this set of equations should also include the possibilities of coupling effects between the various fluxes involved. A simultaneous solution of this set of equations with appropriate initial and boundary conditions is then needed to evaluate the behavior of the particulate system. Unfortunately, the complete analytical solution is not yet available due to mathematical difficulties. Therefore, most of the available mathematical studies have primarily been limited to studies of a *single* particle where only one transport equation for one phase is considered. Recently some work has been directed to transver involving *swarms* of equal size spherical particles [2, 4]. However, it is well known that the particle size as well as the residence time distributions are not uniform and the assumption of uniformity may lead to error [3].

The present study aims to generalize the effects of particle size and residence time

distributions on the transport rates. For the purpose of generality it first analyzes the effect of a general normalized particle size distribution on coupled heat and mass transfer in a multi-component, multiphase particulate system. To arrive at a working approximate description of the system under study we analyze a few simplified examples of moment equations of the distribution function, their experimental verifications and their effect on the rate of mass and heat transfer in particulate systems.

GENERAL FORMULATION

The system considered here is a dispersion or a suspension which consists of a large number of spherical particles (i.e. bubbles, drops or solid particles). The radius of a typical particle is a and the population has a normalized size distribution $f(a)$, so that

$$\int_0^{\infty} f(a) da = 1 \quad (1)$$

where $f(a) da$ represents a fraction in the size range $a \pm da/2$. The particular form of $f(a)$ depends on the physical parameters of the particulate system and the operating conditions. A detailed analysis of $f(a)$ is presented in the second part of this paper.

Introducing linear laws

$$J_i = \sum_j L_{ij} X_j, \quad (i, j = 1, 2, \dots, n-1, T),$$

the independent transport fluxes of a simultaneous (coupled) mass and heat transfer in an n -component particulate system consisting of π phases ($\alpha = 1, 2, \dots, \pi$) can be written as

$$\vec{J}_{Di}^\alpha = - \left\{ \sum_{j=1}^{n-1} \Psi_{ij}^\alpha \text{grad } C_j^\alpha + \Psi_{iT}^\alpha \text{grad } T^\alpha \right\} \quad (i = 1, 2, \dots, n-1) \quad (2)$$

$$\vec{J}_H^\alpha = - \left\{ \sum_{j=1}^{n-1} \Psi_{Tj}^\alpha \text{grad } C_j^\alpha + \Psi_{TT}^\alpha \text{grad } T^\alpha \right\} \quad (3)$$

where the superscript refers to each homogeneous phase α , and the subscripts D and H refer to diffusional and heat flux respectively.

In the following analysis we assume that the volumetric heat capacity $\rho^\alpha C_p^\alpha$ and the coefficients Ψ_{ij}^α are constant for the range of the operating conditions prevailing in the system considered. The nature of Ψ_{ij}^α can be evaluated by the principles of non-equilibrium thermodynamics [1]. For the special case of a binary mixture ($n = 2$), (2) and (3) may take the form

$$\vec{J}_{D1}^\alpha = - \Psi_{11}^\alpha \text{grad } C_1^\alpha - \Psi_{1T}^\alpha \text{grad } T^\alpha \quad (2a)$$

$$\vec{J}_H^\alpha = - \Psi_{T1}^\alpha \text{grad } C_1^\alpha - \Psi_{TT}^\alpha \text{grad } T^\alpha \quad (3a)$$

where

$$\Psi_{11}^\alpha = \frac{L_{11}^\alpha \left(\frac{\partial \mu_1}{\partial C_1} \right)_{P, T, C_2}^\alpha}{C_2^\alpha T^\alpha} \quad (2b)$$

$$\Psi_{1T}^\alpha = L_{1T}^\alpha / (T^\alpha)^2 \quad (2c)$$

$$\Psi_{T1}^\alpha = \frac{L_{T1}^\alpha \left(\frac{\partial \mu_1}{\partial C_1} \right)_{P, T, C_2}^\alpha}{C_2^\alpha T^\alpha} \quad (3b)$$

$$\Psi_{TT}^\alpha = \frac{L_{TT}^\alpha}{(T^\alpha)^2} \quad (3c)$$

$$C_j^\alpha = \frac{\rho_j^\alpha}{\rho^\alpha}; \quad \left(\sum_{j=1}^n C_j^\alpha = 1 \right)$$

and when the L_{ij} are the proportional coefficients of the linear laws expressed in terms of gradients of the chemical potentials μ_j^α and the temperature T^α [1].

From the conservation equations for mass and energy in the absence of external fields and internal sources or sinks,

$$\rho^\alpha \frac{DC_i^\alpha}{Dt} = - \text{div } \vec{J}_{Di}^\alpha; \quad (i = 1, 2, \dots, n-1) \quad (4)$$

$$\rho^\alpha C_p^\alpha \frac{DT^\alpha}{Dt} = - \text{div } \vec{J}_H^\alpha \quad (5)$$

where

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \vec{v} \cdot \text{grad}$$

is the substantial time derivative.

Let

$$\bar{\xi}^\alpha = (C_1^\alpha, C_2^\alpha, \dots, C_{n-1}^\alpha, T_n^\alpha)^T \quad (6)$$

where $()^T$ denotes the transposed n -dimensional vector with components $C_1, C_2, \dots, C_{n-1}, T_n$.

Introducing now the matrix

values of (12) were shown [6] to be given by

$$\lambda_{1,2}^\alpha = \frac{1}{2} \left\{ \Psi_{11}^\alpha / \rho^\alpha + \Psi_{TT}^\alpha / \rho^\alpha C_p^\alpha \pm \left([\Psi_{11}^\alpha / \rho^\alpha - \Psi_{TT}^\alpha / \rho^\alpha C_p^\alpha]^2 + \frac{4\Psi_{1T}^\alpha \cdot \Psi_{T1}^\alpha}{(\rho^\alpha)^2 C_p^\alpha} \right)^{\frac{1}{2}} \right\} \quad (12a)$$

$$\Omega^\alpha = \begin{bmatrix} \Psi_{11}^\alpha / \rho^\alpha & \Psi_{12}^\alpha / \rho^\alpha & \dots & \Psi_{1,n-1}^\alpha / \rho^\alpha & \Psi_{1T}^\alpha / \rho^\alpha \\ \Psi_{21}^\alpha / \rho^\alpha & \Psi_{22}^\alpha / \rho^\alpha & \dots & \Psi_{2,n-1}^\alpha / \rho^\alpha & \Psi_{2T}^\alpha / \rho^\alpha \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ \Psi_{n-1,1}^\alpha / \rho^\alpha & \Psi_{n-1,2}^\alpha / \rho^\alpha & \dots & \Psi_{n-1,n-1}^\alpha / \rho^\alpha & \Psi_{n-1,T}^\alpha / \rho^\alpha \\ \Psi_{T1}^\alpha / \rho^\alpha C_p^\alpha & \Psi_{T2}^\alpha / \rho^\alpha C_p^\alpha & \dots & \Psi_{T,n-1}^\alpha / \rho^\alpha C_p^\alpha & \Psi_{TT}^\alpha / \rho^\alpha C_p^\alpha \end{bmatrix} \quad (7)$$

we find that (4) and (5) can be put in the compact matrix form

$$\frac{D\bar{\xi}^\alpha}{Dt} = \Omega^\alpha \nabla^2 \bar{\xi}^\alpha \quad (8)$$

where, in general, Ω^α is a non-symmetrical matrix whose components off the main diagonal are the coupling coefficients among the various fluxes involved.

Making the transformation

$$\bar{\xi}^\alpha = \mathbf{M}^\alpha \bar{\zeta}^\alpha \quad (9)$$

(8) can be transformed into the *decoupled* equations*

$$\frac{D\bar{\zeta}^\alpha}{Dt} = \Lambda^\alpha \nabla^2 \bar{\zeta}^\alpha \quad (10)$$

where

$$\Lambda^\alpha = [\mathbf{M}^\alpha]^{-1} \Omega^\alpha \mathbf{M}^\alpha \quad (11)$$

is a diagonal matrix with elements, λ_i

$$\Lambda^\alpha = \text{diag}(\lambda_1^\alpha, \lambda_2^\alpha, \dots, \lambda_{n-1}^\alpha, \lambda_n^\alpha). \quad (12)$$

In the case of a binary mixture the eigen-

It follows from the second law of thermodynamics, Onsager relation; $L_{ij} = L_{ji}$, the thermodynamic stability condition (1);

$$(\partial \mu_1 / \partial C_1)_{P,T,C_2} \geq 0$$

and the definitions (2b), (2c), (3b), and (3c), that $\lambda_{1,2}^\alpha$ have real values. Theory and experiment [23, 24] reveal that $\Psi_{11}\Psi_{TT} > \Psi_{1T}\Psi_{T1}$ therefore, $\lambda_{1,2}^\alpha$ are non-negative. Depending on the units and frame of reference of Ψ_{ij}^α and C_j^α and according to the same thermodynamic principles a matrix may be found that reduces Ω^α to a diagonal form, the elements of which are real numbers.

To analyze equations (10) we introduce an integral transformation with a suitable kernel $K^\alpha(\bar{\tau}^\alpha, t)$ defined by

$$\langle \bar{\zeta}^\alpha(\bar{\tau}^\alpha) \rangle = \int_0^\infty K^\alpha(\bar{\tau}^\alpha, t) \bar{\zeta}^\alpha(t) dt \quad (13)$$

so that $\langle \bar{\zeta}^\alpha(t) \rangle$ is a time average of $\bar{\zeta}^\alpha(t)$ over $K^\alpha(\bar{\tau}^\alpha, t)$. The integral operator (13) can be applied to the decoupled equations (10), which

* The diagonalization of Ω^α is feasible since its eigenvalues are distinct.

yield ordinary differential equations. These can then be solved with the appropriate initial and boundary conditions to give the average concentrations and the temperature as well as average transfer fluxes [6]. The advantage of this method is that the kernel $K^a(\bar{\tau}^a, t)$ can be chosen to be the residence time distribution function for the particles as well as for the continuous phase, so that $\langle \bar{\zeta}^a(\bar{\tau}^a) \rangle$ becomes the expected value of $\rho^a(t)$, and $\bar{\tau}^a$ can be interpreted as a parameter of the residence time distribution in phase α . Some other mathematical advantages of this method will be discussed later.

In the general case of swarms of spherical bubbles, drops or solid particles, the $\bar{\zeta}^a$ field is expected to be spherically symmetric when the origin of coordinates is fixed on the center of mass of a typical particle. Therefore by using spherical coordinates and the initial condition:

$$t = 0, r \neq a, \bar{\zeta}^a = \bar{\zeta}_0^a \quad (14)$$

i.e.

$$t = 0, r \neq a, C_j^a = C_{j0}^a (j = 1, 2, \dots, n-1) \quad (14a)$$

$$t = 0, r \neq a, T^a = T_0^a \quad (14b)$$

the integral operators (13) can be applied to equations (10) to yield ordinary *decoupled* differential equations

$$\lambda_i \nabla^2 \langle \zeta_i^a \rangle = (\nabla^a \cdot \nabla) \langle \zeta_i^a \rangle + \int_0^\infty K^a(\bar{\tau}^a, t) \frac{\partial \zeta_i^a}{\partial t} dt \quad (15)$$

independent of the time variable. Using appropriate boundary conditions (which should include the average distance between adjacent particles [2-4, 10], equilibrium relationships between interfacial concentrations and temperatures, in addition to equality of mass- and heat-transfer fluxes on both sides of the interfaces), equation (15) can be solved in principle to yield $\langle \zeta_i^a \rangle$ as a function of space coordinates. However, in the case of dispersions or suspensions of swarms of particles, the question

arises as to the functional form of $\bar{\nabla}^a$ —a quantity which is very difficult to evaluate by experimental or theoretical methods [25]. Nevertheless, the convective term in (15) can be assumed negligible according to physical arguments presented in previous works [4, 6]. This assumption is a good approximation in the domain of large number of particles per unit volume, presence of surface-active agents and relatively small particle sizes whereby the particles are completely entrained by the eddies of the continuous phase [4, 10-12]. Consequently, in systems of constant density (isotropic mixtures, or mass diluted systems, i.e. $C_j^a \ll 1$ for $j = 1, 2, \dots, n-1$ and $C_n^a \cong 1$) which can also be classified in the above mentioned domain, the convective contribution to the total transfer rate can be neglected, thereby resulting in a considerable simplification in the solution of equations (15).

In a two-phase system the contribution to the total interfacial transfer rate from a population of particles in a size range $a, \pm da/2$ is then calculated from

$$d\bar{W}(\xi) = N4\pi a^2 \langle \bar{J}(a) \rangle f(a) da \quad (16)$$

where

$$\begin{aligned} \langle \bar{J}(a) \rangle &= \langle \bar{J}^a(a) \rangle = -\Omega^a \left(\frac{\partial \langle \bar{\zeta}^a \rangle}{\partial r} \right)_{r=a} \\ &= -M^a A^a \left(\frac{\partial \langle \bar{\zeta}^a \rangle}{\partial r} \right)_{r=a} \end{aligned} \quad (17)$$

and

$$N = 3\Phi V / 4\pi(\bar{a}_3)^3 \quad (18)$$

is the total number of particles in the dispersion (or suspension) whose volume V , Φ is the volumetric dispersed-phase holdup fraction, and \bar{a}_3 is the mean volume radius of the particle population defined by

$$\bar{a}_3 = \left(\int_0^\infty a^3 f(a) da \right)^{1/3} \quad (19)$$

Here $\bar{J}(a, t)$ is the instantaneous interfacial flux. Therefore, the expected value $\langle \bar{J}(a) \rangle$ takes into account the variations in residence time among the entire particle population.

Summing up the contribution from all size ranges of the particle population gives the total average heat or mass transfer, $\bar{W}(\xi)$ in the particulate system, i.e.

$$\begin{aligned}\bar{W}(\xi) &= \frac{3\Phi V}{(\bar{a}_3)^3} \int_0^\infty a^2 f(a) \int_0^\infty K^a(\bar{\tau}^a, t) \bar{J}^a(a, t) dt da \\ &= -M^a A^a \left\{ \frac{3\Phi V}{(\bar{a}_3)^3} \int_0^\infty a^2 f(a) \left(\frac{\partial \langle \bar{\xi}^a \rangle}{\partial r} \right)_{r=a} da \right\}\end{aligned}\quad (20)$$

Hence the quantity

$$I_{\text{ensemble}}^a = -\Omega^a \frac{3\Phi V}{(\bar{a}_3)^3} \int_0^\infty \int_0^\infty K^a(\bar{\tau}^a, t) a^2 f(a) \{ \dots \} dt da \quad (21)$$

can be viewed as a general integral operator for a particle population, which yields the total average interfacial transfer rate when operated on local instantaneous gradients at the interface. Apparently this method can be extended to cases involving simultaneous heat and mass transfer in reaction systems, simultaneous mass, heat, and electric charge transfer, etc. [1]. A concentric spherical model [2] with contact and mixing mechanism has been employed for analytical evaluation of $\langle \bar{\xi}^a \rangle$, and $\bar{W}(\xi)$ in terms of a general particle size distribution and the cases of coupled heat and mass transfer in a binary two-phase particulate system [6] and simultaneous mass (or heat) transfer with zeroth and/or first order irreversible chemical reaction in a binary two-phase particulate system [10]. Further evaluations depend on a more explicit description of moment equations of the distribution functions.

MOMENT EQUATIONS

Moment equations of particle size distribution can be used to characterize the particle population by evaluating such quantities as cumulative number density, cumulative interfacial area, cumulative volume, interrelationships among

the various mean sizes of the population and the effects of size distribution on the transfer fluxes involved.

A general statistical mechanical approach to the moment equations has been recently formulated by Hulburt and Katz [5]. They describe a typical particle in terms of phase-space coordinates that can be separated into external coordinates (specifying the location of the particle in the processing unit) and internal coordinates (specifying particle size and other aspects of the particle quality as may be relevant).

An analogous continuity equation for particulate systems, based on the particles in the phase-space, led Hulburt and Katz to the general equation

$$\begin{aligned}\frac{\partial f(x_i, r_j, t)}{\partial t} + \sum_{i=1}^3 \frac{\partial \{V_i(x_i, t) f(x_i, r_j, t)\}}{\partial x_i} \\ + \sum_j \frac{\partial \{G_j[C(x_i, t), T(x_i, t), r_j] f(x_i, r_j, t)\}}{\partial r_j} \\ = B[C(x_i, t), T(x_i, t), r_j]\end{aligned}\quad (22)$$

where x_i and r_j are the external and internal coordinates respectively, $f(x_i, r_j, t)$ number density of particles in the system at time t with phase coordinates x_i and r_j , $V_i(x_i, t)$, $C(x_i, t)$ and $T(x_i, t)$ are, respectively, velocity, concentration and temperature, G_j is the growth rate function and B is the nucleation function (which includes the rate of formation of new particles by nucleation, agglomeration, breakage, feed and take-off mechanisms). If G_j and B are known functions of C , T , r_j , and the operating conditions then (22) can be solved, in principle, simultaneously with the bulk transport equations for mass, energy, and momentum (and suitable initial and boundary conditions) to give $f(x_i, r_j, t)$, $C(x_i, t)$, $T(x_i, t)$, $V_i(x_i, t)$. If coupling effects among the various transport processes involved are taken also into account then these solutions can furnish the complete mathematical characterization of the system. Apparently this

is not yet possible and we assume here that the particle number density depends only on one internal coordinate, a , the typical size of a population of spherical particles. This assumption means that at a given time t , G_j and B are the same in any location in the particulate system so that $f(x_i, r_j, t)$ reduces to $f(r_1 = a, \beta, t)$ where β is a parameter of the particle size distribution that depends on the solution of equation (22) or on experimental evaluations. We define now the n th moment of the particle size distribution μ_n by

$$\mu_n(\beta, t) = \int a^n f(a, \beta, t) da; n = 1, 2, \dots \quad (23)$$

where the integration extends over all possible values of a .

For most purposes, as will be shown later, the integration can be extended from 0 to ∞ without introducing any significant error. Therefore, the following quantities can be defined for the dispersed phase:

Cumulative number density, per cent

$$= 100 \int_0^a f(a, \beta, t) da / \mu_0(\beta, t) \quad (24)$$

Cumulative interfacial area, per cent

$$= 100 \int_0^a a^2 f(a, \beta, t) da / \mu_2(\beta, t) \quad (25)$$

Cumulative volume, per cent

$$= 100 \int_0^a a^3 f(a, \beta, t) da / \mu_3(\beta, t) \quad (26)$$

where $f(a, \beta, t)$ is a normalized distribution function.

In addition, the following mean sizes can be defined:

Surface mean radius (Sauter mean):

$$\bar{a}_{32} = \frac{\sum_i n_i a_i^3}{\sum_i n_i a_i^2} \equiv \frac{\mu_3(\beta, t)}{\mu_2(\beta, t)} \quad (27)$$

Mean volume radius:

$$\bar{a}_3 = \left(\frac{\sum_i n_i a_i^3}{\sum_i n_i} \right)^{\frac{1}{3}} \equiv \left[\frac{\mu_3(\beta, t)}{\mu_0(\beta, t)} \right]^{\frac{1}{3}} \quad (28)$$

Volume mean radius:

$$\bar{a}_{43} = \frac{\sum_i n_i a_i^4}{\sum_i n_i a_i^3} \equiv \frac{\mu_4(\beta, t)}{\mu_3(\beta, t)} \quad (29)$$

Mean surface radius:

$$\bar{a}_2 = \left(\frac{\sum_i n_i a_i^2}{\sum_i n_i} \right)^{\frac{1}{2}} \equiv \left[\frac{\mu_2(\beta, t)}{\mu_0(\beta, t)} \right]^{\frac{1}{2}} \quad (30)$$

Interrelationships among these means can be evaluated when $f(a, \beta, t)$ is known. For example, many investigators report experimental data in terms of either a_{32} or a_3 . To compare these data one should know the ratio

$$\frac{\bar{a}_{32}}{\bar{a}_3} = \frac{[\mu_3(\beta, t)]^{\frac{1}{3}}}{\mu_2(\beta, t)} \quad (31)$$

since $\mu_0(\beta, t) = 1$.

The dispersed-phase holdup fraction can now be expressed as

$$\Phi = \frac{4}{3} \pi N_v \mu_3(\beta, t) \quad (32)$$

where N_v is the total number of particles per unit volume of the particulate system (i.e. $N_v = N/V$).

A differential interfacial area of the entire particle population can be expressed as

$$dA = 4\pi V N_v a^2 f(a, \beta, t) da \quad (33)$$

Hence if the instantaneous rate flux $\bar{J}(a, t)$ is given, then the instantaneous total flux in the system can be evaluated from

$$\bar{W}(t) = 4\pi V N_v \int \bar{J}(a, t) a^2 f(a, \beta, t) da \quad (34)$$

where the integration extends over all possible values of the particle size. Let us consider first a well-mixed continuous inflow-outflow particulate system with a time invariant inventory of N particles, delivered into the vessel ω times

per unit time. In a *well-mixed* vessel, the particle bulk environmental coordinates are uniform and under these conditions $f(x_b, r_p, t)$ is completely characterized by a single internal coordinate τ , the time a particle in a feed group of injected tracer elements has spent in the vessel. Hulburt and Katz [5] show that for this case $D\tau/Dt = 1$ where t is the clock time and the phase-space coordinates become the τ -axis, i.e. the number density of particles in the phase-space becomes $f(\tau, t) d\tau$ equal to the number of particles in the system at time t with age in the vessel in the range $\tau \pm d\tau/2$. This is equal to the residence time distribution in the take-off stream as well as to the age distribution in the vessel as a whole.

Under these conditions B represents the net inflow-outflow particle balance which can be expressed in terms of Dirac delta function $\delta(\tau)$ [5]:

$$B(\omega, \tau, t) = \omega[N\delta(\tau) - f(\tau, t)] \quad (35)$$

where $\omega N[\delta(\tau)]$ represents the rate of intake of particles, all at age zero and $\omega f(\tau, t)$ represents the rate of take-off. Under these conditions (22) becomes

$$\frac{\partial f(\tau, t)}{\partial t} + \frac{\partial [f(\tau, t)]}{\partial \tau} = \omega[N\delta(\tau) - f(\tau, t)]. \quad (36)$$

Hulburt and Katz [5] solved (36) explicitly to give

$$f(\tau, t) = \exp(-\omega t) f(\tau - t, 0) + N\omega \exp(-\omega \tau) [\Delta(\tau) - \Delta(\tau - t)] \quad (37)$$

where

$$\Delta(\tau) = \begin{cases} 0 & \tau < 0 \\ 1 & \tau > 0 \end{cases} \quad (38)$$

is the Heaviside unit step function whose time derivative is $\delta(\tau)$. Equation (37) shows how $f(\tau, t)$ evolves toward its well-known steady-state solution for a well-mixed vessel

$$\lim_{t \rightarrow \infty} f(\tau, t) = N\omega \exp(-\omega \tau) \Delta(\tau). \quad (39)$$

Consequently, for the case of transfer in a well-

mixed vessel the kernel $K^a(\bar{\tau}^a, t)$ in equation (21) can be chosen as

$$K^a(\bar{\tau}^a, t) = [\bar{\tau}^a]^{-1} \exp(-t/\bar{\tau}^a) \quad (40)$$

where we recall also that t in equation (13) is actually τ , the time a particle has spent in the vessel and $\bar{\tau}^a$ is the holding time of either phase. Frequently $\bar{\tau}^a$ is the same for both phases as observed by Schmidz and Amundson [7] and Trambouze [13] when there is no obstruction of the flow of either phase in the exit. Hence, for this case the holding time is also the average residence time.

The integral transformation (13) with kernel (40) is seen to be accomplished by taking Laplace transforms of equations (10) with $\exp(-st)$, where $s = 1/\bar{\tau}^a$ and dividing the transformed quantity by $\bar{\tau}^a$. Hence, the expected value of $\langle \bar{J}(a) \rangle$ is simply given by

$$\langle \bar{J}(a) \rangle = -\frac{1}{\bar{\tau}^a} \Omega^a \left(\frac{\partial [\bar{\xi}^a(s)]_{s=1/\bar{\tau}^a}}{\partial r} \right)_{r=a} \quad (41)$$

i.e. the tedious (and usually impossible) inverse transform of the transformed partial differential equations (10) becomes an unnecessary procedure thereby resulting in a significant mathematical simplification.

For any general case of residence time distribution in any *real* system we can choose $K^a(\bar{\tau}^a, t)$ so that

$$K^a(\bar{\tau}^a, t) = [\bar{\tau}^a]^{-1} \exp \left[-\eta^a \left(\frac{t - \varepsilon^a}{\bar{\tau}^a} \right) \right] \quad (42)$$

where $\exp[-\eta^a(t - \varepsilon^a)/\bar{\tau}^a]$ is the general residence time distribution proposed by Wolf and Resnick [14] for real systems. In their distribution η is a measure of the efficiency of mixing and ε is a measure of the phase shift in the system. For well-mixed vessel $\eta = 1$ and $\varepsilon = 0$, whereas for plug flow, η tends to infinity. The behavior of η and ε for combinations of well-mixed vessel with plug flow, dead space, short circuiting, and system lag is shown in Table 1. The data of Wolf and Resnick indicate that such systems as flotation cells, fluidized beds and

stirred reactors are essentially described by $\eta^\alpha \doteq 1$, $\varepsilon^\alpha/\bar{\tau}^\alpha \doteq 0$ whereas for gas flow in a pipe $\eta \gg 1$ and $\varepsilon/\bar{\tau} \rightarrow 1$. Assuming $\bar{\tau}^\alpha$, η^α and ε^α are known experimentally for a given particulate system under study, we can now re-express the

Table 1. Properties of the distribution function (42), [14]

	η	$\varepsilon/\bar{\tau}$
Well-mixed Vessel	1	0
with plug flow	>1	>0
with dead space	>1	0
with short circuiting	<1	<0
with error in $\bar{\tau}$	≥ 1	0
with system lag	1	>0

integral operator $I_{\text{ensemble}}^\alpha$ in terms of a modified Laplace transform divided by a characteristic time parameter of the system $\bar{\tau}_*^\alpha$ so that

$$K^\alpha(\bar{\tau}^\alpha, \eta^\alpha, \varepsilon^\alpha, t) = [\bar{\tau}_*^\alpha]^{-1} \exp(-s^\alpha t) \quad (43)$$

where

$$\hat{\tau}^\alpha = \frac{\bar{\tau}^\alpha}{\eta^\alpha} = \frac{1}{s^\alpha} \quad (43a)$$

is the modified average residence time defined by

$$\hat{\tau}^\alpha = \frac{\int_0^\infty t \exp(-s^\alpha t) dt / \bar{\tau}_*^\alpha}{\int_0^\infty \exp(-s^\alpha t) dt / \bar{\tau}_*^\alpha} = \frac{\bar{\tau}^\alpha}{\eta^\alpha} \quad (44)$$

and $\bar{\tau}_*^\alpha$ a characteristic time parameter of the system:

$$\bar{\tau}_*^\alpha = \bar{\tau}^\alpha \exp[-\eta^\alpha \varepsilon^\alpha / \bar{\tau}^\alpha]. \quad (45)$$

Thus, using the modified Kernel (43) the set of transport equations (10) can be solved for any real system and again the inverse Laplace transformation becomes an unnecessary procedure. This removes a considerable mathematical difficulty in obtaining analytical solutions for multicomponent mass and heat transfer in real multiphase particulate systems.

EFFECT OF PARTICLE SIZE DISTRIBUTION ON TRANSPORT RATE

In this section we consider a useful particle size distribution that can be substituted in equation (20) to evaluate the total average transfer rate $\bar{W}(\bar{\xi})$. Such one is the particle size distribution that has been proposed recently by Bayens [15] for hydrosols coagulating in Brownian motion:

$$f(a, N_v, \Phi) = 4 \left(\frac{\alpha^3}{\pi} \right)^{\frac{1}{2}} a^2 \exp[-\alpha a^2] \quad (46)$$

where

$$\alpha = \left(\frac{16(\sqrt{\pi}N_v)}{3\Phi} \right)^{\frac{1}{2}} > 0 \quad (47)$$

(46) is a normalized function of the same form as the Maxwell-Boltzmann speed distribution for gaseous atoms. It has been checked for a batch system in which α changes with time because the coagulation rate changes the ratio N_v/Φ in time. The agreement of this equation with experimental data of Swift and Friedlander [16] is excellent [15]. For semibatch system (continuous inflow-outflow of the dispersed phase with simultaneous transfer and sink rates) and for continuous systems where both phases enter and leave the system under constant operating conditions $G(a, t)$, $B(a, t)$, $C(a, t)$, $T(a, t)$, $V(a, t)$, eventually become time invariants (steady state) thereby resulting in a time invariant particle size distribution. Consequently, the rates of inflow-outflow, turbulence, diffusion, and heat transfer (with or without sources), nucleation, agglomeration (coalescence) and breakage of particles reach a steady-state value with a constant N_v/Φ ratio. The ratio N_v/Φ is constant (but different) for each steady state.

Hence, by substituting (18) in (47), (46) is re-expressed as

$$f(a, \bar{a}_3) = \frac{16}{\pi(\bar{a}_3)^3} a^2 \exp \left[- \left(\frac{4}{\sqrt{(\pi)(\bar{a}_3)^3}} \right)^{\frac{1}{2}} a^2 \right] \quad (48)$$

which indicates that the entire particle size distribution may be determined if only one parameter, \bar{a}_3 is known as a function of the physical properties of the system and the operating variables. For a batch system \bar{a}_3 changes with time of operation due to agglomeration, etc. For semibatch and continuous systems \bar{a}_3 is time invariant under steady-state conditions. Consequently (48) can be checked against experimental data of, say, gas-liquid dispersions. This can be done by measuring cumulative number density and cumulative volume percents.

Bringing (48) to (24), (25), and (26), and carrying the appropriate integrations finally gives:

Cumulative number density, per cent

$$= 100 \{ \text{erf} [\sqrt{(\alpha)a}] - \alpha \sqrt{(2/\pi)} a \exp [-\alpha a^2] \} \\ = 100 \mu_0^a \quad (49)$$

Cumulative interfacial area, per cent

$$= \frac{100}{\mu_2} \left\{ \frac{3}{2\alpha} \mu_0^a - \frac{2}{\sqrt{(\pi)}} \alpha^{\frac{1}{2}} a^3 \exp [-\alpha a^2] \right\} \quad (50)$$

Cumulative volume, per cent

$$= 100 \left\{ 1 - \left[1 + \alpha a^2 + \frac{\alpha^2}{2} a^4 \right] \exp [-\alpha a^2] \right\} \quad (51)$$

where

$$\alpha = \left(\frac{4}{\sqrt{(\pi)} (\bar{a}_3)^3} \right)^{\frac{1}{2}} \quad (52)$$

and μ_n^a means the n th moment equation when the integration is from zero to a . From (48) the n th moment can be expressed as

$$\mu_n^a = \frac{n+1}{2\alpha} \mu_{n-2}^a - \frac{2}{\sqrt{(\pi)}} \alpha^{\frac{1}{2}} a^{n+1} \exp [-\alpha a^2] \quad (53)$$

for $n \geq 2$.

Introducing the variables

$$\lambda = \alpha^{\frac{1}{2}} \quad (54)$$

$$\sigma = \lambda^2 a^2 \quad (55)$$

$$B^* = \frac{4}{\sqrt{(\pi)}} \alpha^{\frac{1}{2}} \quad (56)$$

and using the tabulated incomplete gamma function (17)

$$\gamma(x, n) = \int_0^x e^{-\sigma} \sigma^{n-1} d\sigma = P(\chi^2 | \nu) \cdot \Gamma(n); \nu = 2n \\ \chi^2 = x \quad (57)$$

when $n \geq 1$, $P(\chi^2 | \nu) \sim P(\chi')$ where

$$x' = \frac{x^2 - \nu}{\sqrt{(2\nu)}} \quad (57)$$

(57) gives

$$\mu_{n-2}^a = B^* \int_0^a a^n \exp (-\lambda^2 a^2) da \\ = \frac{2}{\sqrt{(\pi)} \lambda^{n-2}} \gamma \left(\lambda^2 a^2, \frac{n+1}{2} \right) \quad (58)$$

When $\lambda^2 \rightarrow \infty$ the incomplete gamma function becomes the complete gamma function that can be reduced to the fundamental probability distribution function $P(x)$. Equation (58) may be re-expressed as a confluent hypergeometric function, recurrence formulas and in asymptotic expansions (17).

Equations (49) and (51) were checked against experimental data for gas-liquid dispersion (Figs. 1-5). The disperser and the experimental techniques were described in a previous work [18]. Substituting from (48) and (31) we obtain

$$\bar{a}_{32} = \frac{\Gamma(3.0)}{\Gamma(2\frac{1}{2}) \alpha^{\frac{1}{2}}} = 1.148 \bar{a}_3 \quad (59)$$

The average experimental value of a_{32}/a_3 for air-tap water system was found to be 1.195 whereas that for an air-distilled water system was 1.133, both in good agreement with (59). The agreement between the experimental and predicted values in Figs. 1-5 is reasonable. The deviations observed may result from distortion

of the bubbles from spherical shape, departure from random collisions, coalescence and break-up, and (particularly at the small size range) as a result of experimental error in analyzing the photographs and measuring the size of extremely small particles.

The application of (48) and its various moment equations depends on the knowledge of \bar{a}_3 which is a function of the physical properties and the operating variables of the system. In the case of bubbles and drops the average size is characterized by the Weber number. Shinnar and Church [19] developed some theoretical predictions for average particle size by using Kolmogoroff's theory of local isotropy. For breakage as the dominant mechanism and at low Φ values, they obtained

$$\bar{a}_{32} = B_1 L (N_{We})^{-0.6} = B_1 L \left(\frac{L^3 \rho (N')^2}{\gamma^*} \right)^{-0.6} \quad (60)$$

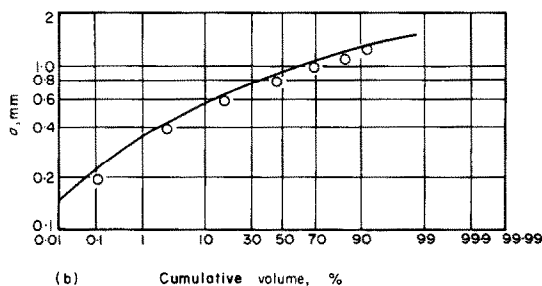
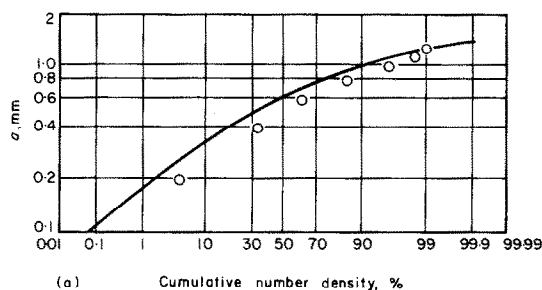


FIG. 1. Air-tap water dispersion; $N' = 1285$ rev/min; $\bar{a}_3 = 0.737$ mm; $\bar{a}_{32}/\bar{a}_3 = 1.149$. Solid lines represent equation (49) in Fig. 1(a) and equation (51) in Fig. 1(b).

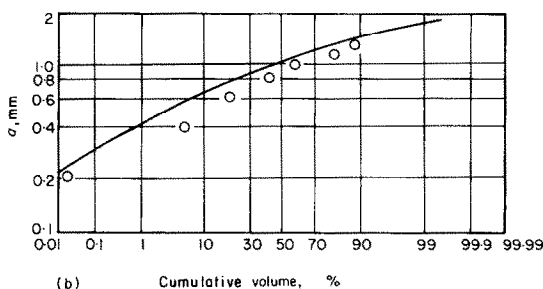
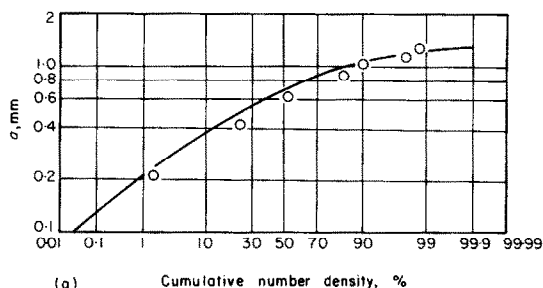


FIG. 2. Air-tap water dispersion; $N' = 1460$ rev/min; $\bar{a}_3 = 0.742$ mm; $\bar{a}_{32}/\bar{a}_3 = 1.242$. Solid lines represent equation (49) in Fig. 2(a) and equation (51) in Fig. 2(b).

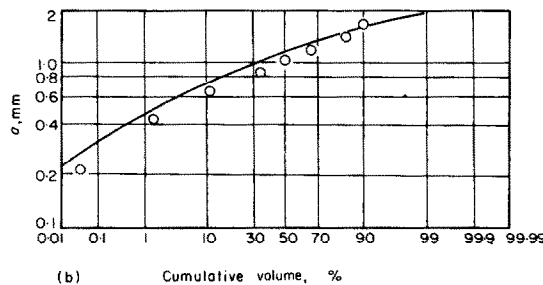
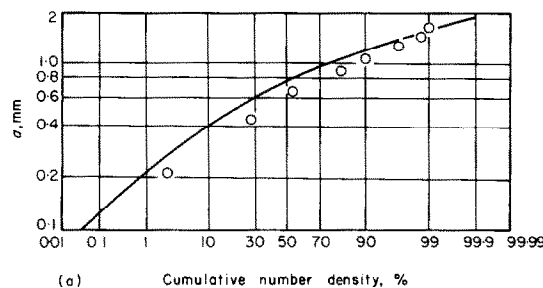


FIG. 3. Air-distilled water dispersion; $N' = 1080$ rev/min; $\bar{a}_3 = 0.929$ mm; $\bar{a}_{32}/\bar{a}_3 = 1.158$. Solid lines represent equation (49) in Fig. 3(a) and equation (51) in Fig. 3(b).

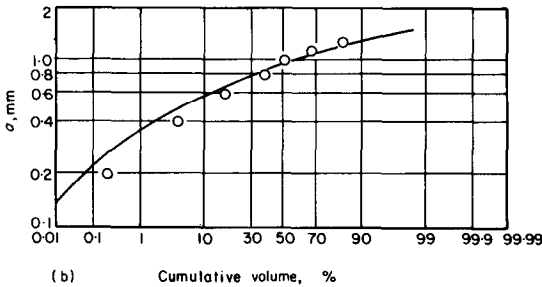
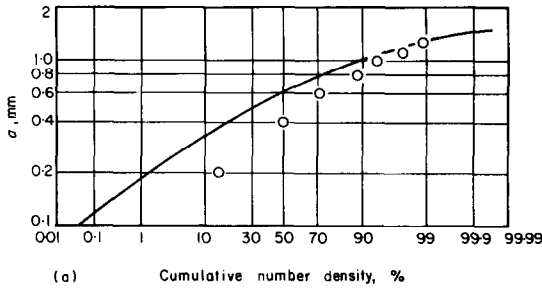


FIG. 4. Air-distilled water dispersion; $N' = 1200$ rev/min; $\bar{a}_3 = 0.850$ mm; $\bar{a}_{32}/\bar{a}_3 = 1.104$. Solid lines represent equation (49) in Fig. 4(a) and equation (51) in Fig. 4 (b).

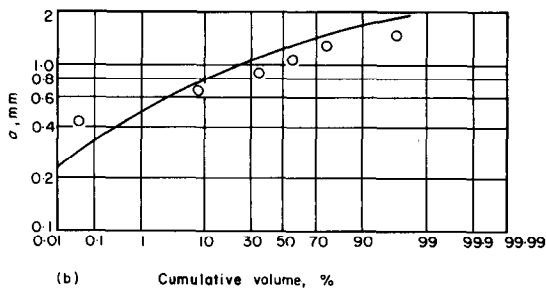
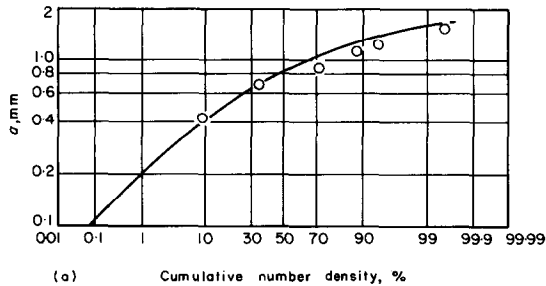


FIG. 5. Air-distilled water dispersions; $N' = 1300$ rev/min; $\bar{a}_3 = 1.012$ mm; $\bar{a}_{32}/\bar{a}_3 = 1.105$. Solid lines represent equation (49) in Fig. 5(a) and equation (51) in Fig. 5(b).

where L is a linear dimension of the impeller, N' , its rotational speed, γ^* the interfacial tension and B_1 a constant. This theoretical result is in good agreement with the experimental results of Calderbank [21]

$$\bar{a}_{32} = B_2(1 + 9\Phi) L(N_{we})^{-0.6} \quad (61)$$

and Chen and Middleman [20]

$$\bar{a}_{32} = B_3 L(N_{we})^{-0.6} \quad (62)$$

at very low values of Φ . For immiscible liquid-liquid dispersions Chen and Middleman obtained $B_1 = 0.026$ for very small values of Φ wherein coalescence of droplets plays no role in the dispersion mechanism. The experimental results of Vermeulen *et al.* [22] (for liquid-liquid and gas-liquid dispersions)

$$\bar{a}_{32} = B_4 \Phi(N_{we})^{-0.6} \quad (63)$$

support also, in general, the form of (61).

Substituting (61) in (59) and then in (48) gives the general dependency of $f(a, a_3)$ and its moment equations on γ^* , ρ , N' , L and Φ : i.e. α in equation (46) and in the expressions for μ_n^a should be given by

$$\alpha = \frac{(4^{\frac{1}{3}} \cdot 1.318/\pi^{\frac{1}{3}})(N_{we})^{1.2}}{B_2^2(1 + 9\Phi)^2 L^2} > 0. \quad (64)$$

The total average heat and/or mass transfer in a gas-liquid or liquid-liquid dispersion can thus be obtained from

$$\begin{aligned} \bar{W}(\bar{\xi}) = & -\bar{M}^a A^a \left\{ \frac{4.539 \Phi V(N_{we})^{-1.8}}{B_2^3(1 + 9\Phi)^3 L^3} \right. \\ & \times \left. 4 \left(\frac{\alpha^3}{\pi} \right)^{\frac{1}{3}} \int_0^{\infty} a^4 \exp(-\alpha a^2) \left(\frac{\partial \langle \bar{\xi}^a \rangle}{\partial r} \right)_{r=a} da \right\} \end{aligned} \quad (65)$$

where α is given by (64).

Using the modified average residence time (45) with the Laplace transform in terms of Kernel (43) gives

$$\left(\frac{\partial \langle \bar{\xi}^a \rangle}{\partial r} \right)_{r=a} = \left[\frac{\partial \bar{\xi}^a(s^a)}{\partial r} \right]_{s^a = \eta^a/\bar{\tau}^a} \quad (66)$$

where $\bar{\zeta}^\alpha(s^\alpha) = \bar{\zeta}^\alpha\{r\}$ is the solution of (15) with a given set of initial and boundary conditions. Since the inverse transform of $\bar{\zeta}^\alpha(s^\alpha)$ becomes unnecessary, the procedure is rather simple.

The practical advantages of expressing α in terms of Φ , L and the Weber number [equation (64)] are two:

(a) Variations in the moment equations as functions of the independent variables N' and L can be easily formulated and checked against experimental data.

(b) The effects of changes of the independent operating parameters N' and L on $\bar{W}(\bar{\xi})$ can be obtained.

A detailed study of these effects is now under progress and the results will be reported in due time.

ACKNOWLEDGEMENT

The author would like to express his sincere gratitude to the School of Engineering of the University of Pittsburgh and, in particular, to Dean H. E. Hoelscher and Professor E. B. Stuart for the general support given for conducting this research.

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Résumé—Une théorie est présentée pour le cas général du transport couplé de chaleur et de masse à plusieurs constituants dans des dispersions et des suspensions avec des distributions de temps de séjour et des tailles de particules. La formulation générale décrit le comportement de systèmes industriels et biologiques de particules à l'aide d'équations connues à partir de la thermodynamique en non-équilibre

et des phénomènes de transport. En employant la notation matricielle et des transformations appropriées, les équations aux dérivées partielles qui en découlent sont d'abord découplées et transformées alors en équations différentielles ordinaires en utilisant un opérateur intégral dont le noyau tient compte des distributions de tailles de particules et de temps de séjour. Les équations aux moments de la distribution des tailles sont formulées et employées pour évaluer les relations générales entre les tailles moyennes, la surface interfaciale, la fraction suspendue de la phase dispersée, et les taux moyens totaux de transport, à partir de la population des particules. Les taux moyens totaux de transport de chaleur et de masse dans les systèmes multiphasés de particules sont évalués en fonction d'une distribution générale normalisée des tailles et des valeurs attendues des différents flux. Les propriétés d'une distribution générale normalisée des tailles sont discutées et comparées avec les résultats expérimentaux des distributions des tailles de bulles dans une dispersion gaz-liquide.

Zusammenfassung—Es wird eine Theorie für den allgemeinen Fall gekoppelten Wärme- und Vielkomponentenstofftransports in Dispersionen und Suspensionen mit Verweilzeit- und Teilchengrösseverteilungen angegeben. Die allgemeinen Formulierungen beschreiben das Verhalten industrieller oder biologischer Systeme einzelner Teilchen mit Hilfe von Gleichungen, die aus der Nichtgleichgewichtsthermodynamik und aus dem Gebiet der Transportphänomene bekannt sind. Mit Hilfe der Matrizenrechnung und geeigneter Transformationen werden die sich ergebenden Differentialgleichungen zunächst entkoppelt und dann mit einem Integraloperator, dessen Kern die Verteilung der Teilchengrösse und der Verweilzeit berücksichtigt in gewöhnliche Differentialgleichungen umgewandelt. Es werden Gleichungen für die Grössenverteilung formuliert und dazu verwendet, allgemeine Beziehungen zwischen durchschnittlichen Teilchengrössen, Grenzflächengrössen, Verweilzeit in der dispergierten Phase und den gesamten durchschnittlichen Anteilen des Transports von den Teilchen auszuwerten.

Die gesamten durchschnittlichen Anteile des Wärme- und Stoffübergangs in mehrphasigen Teilchensystemen werden mit Hilfe geschätzter Werte für die verschiedenen Flüsse in Form von Ausdrücken einer allgemein genormten Grössenverteilung ausgewertet. Die Eigenschaften einer vorgeschlagenen allgemein genormten Grössenverteilung werden diskutiert und mit Versuchsergebnissen der Blasengrössenverteilung in einer Gas-Flüssigkeitsdispersion verglichen.

Аннотация—Предложена теория для общего случая взаимосвязанных процессов переноса тепла и многокомпонентной массы в дисперсиях и суспензиях при известном распределении времени пребывания и размера частиц. Общие формулировки описывают поведение промышленных или биологических систем частиц с помощью уравнений, обычных для неравновесной термодинамики и описания явлений переноса. При использовании матричного исчисления и соответствующих преобразований результирующие дифференциальные уравнения в частных производных сначала развешиваются, а затем превращаются в обыкновенные дифференциальные уравнения с помощью интегрального оператора, ядро которого учитывает распределение частиц по размеру и времени пребывания. Составлены уравнения сохранения количества движения распределения по размерам, которые используются для расчета общих взаимозависимостей между средними размерами, площадью поверхности раздела, относительной концентрацией дисперсной фазы и полными средними скоростями переноса от комплекса частиц. Полные средние скорости переноса тепла и массы в многофазных системах частиц рассчитываются в виде общего нормализованного распределения по размерам и ожидаемых значений различных потоков. Обсуждаются свойства предлагаемого общего нормализованного распределения по размеру и сравниваются с экспериментальными данными о распределении по размеру пузырьков в газо-жидкостных дисперсиях.