k_L = liquid-phase mass transfer coefficient, m./hr.
p = partial pressure of oxygen in the gas phase, atm.
U_B = average rising velocity of gas bubbles, m./hr.
U_S = superficial gas velocity, m./hr.
Z_F = height of aerated liquid, cm.
Z_L = height of clear liquid, cm.
ϵ_G = average fractional gas holdup, dimensionless
θ = time, hr.

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Heat Transfer Characteristics of Concurrent Gas-Liquid Flow in Packed Beds

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The past decade has seen a proliferation of processes in which gas and liquid react in two-phase flow through vessels packed with catalyst particles. In order to ensure adequate throughput, most of these reactors operate with concurrent flow as opposed to countercurrent flow, for which throughput is severely restricted by flooding. Since these two-phase reaction processes are very often exothermic (that is hydrogenation and oxidation), it is necessary to transfer heat from the process. At the high temperatures, high pressures, and corrosive conditions commonly encountered in such two-phase catalytic reactions, the necessary heat-exchange surface may represent a substantial portion of the reactor section cost. When large heats of reaction must be transferred, the catalyst bed will commonly be split into separate vessels with interstage heat exchangers for cooling. Because of the high cost of such heat-exchanger surface, it might be economically more desirable to exchange heat directly in the reactor, provided the necessary design data were available.

It would appear that careful design is required in gas-liquid reactors to ensure proper temperature control and to reduce the capital cost of the equipment. Few, if any, serious attempts have been made to study heat transfer from packed beds operating with gas-liquid flow. In fact only one paper (*1*) has been published dealing with pressure drop and liquid holdup over a wide range of concurrent, two-phase, packed-bed flow conditions. In the present study, experimental data were obtained for heat transfer to beds, packed with various sizes of spheres, through which the two-phase air-water system was flowing concurrently.

APPARATUS

The flow equipment used in the experimental program was described in a previous paper (*10*) and also in detail elsewhere (*2*). The heat transfer section was attached as an integral part of the packed column and consisted of machined pure copper tube surrounded by a steam chest. Thermocouples were emplaced in this copper wall at various axial and radial positions to enable the average wall temperature to be calculated. All thermocouples were calibrated against an N.B.S. standard thermometer. Steam could be introduced at different positions in the chest to help flatten any temperature profile in the copper wall.

An exit mixing device was designed (*2*) to provide an accurate mixing cup temperature of the outlet gas and liquid stream. It consisted of alternating donut types of baffles followed by a short packed section. Thermocouples were located at various radii inside the small packed section.

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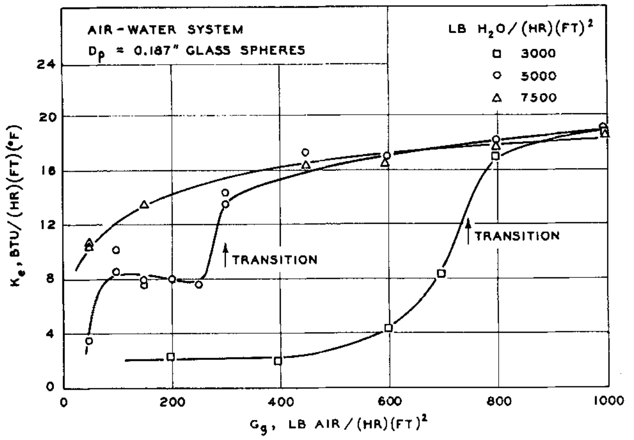

Fig. 1. Effect of gas rate on effective thermal conductivity.

This packing served the dual purpose of inducing further mixing and providing an effective radiation shield for the thermocouples. In practice, this mixing cup worked very well, and under most operating conditions the mixing-cup thermocouples agreed within ± 0.7°F. The inlet bulk temperature, outlet bulk (mixing cup) temperature, and the wall temperature were calculated by averaging the appropriate readings. The maximum deviation for the wall thermocouples was ± 4.0°F. from the average temperature.

To ensure a constant inlet temperature during the heat transfer runs, the fluid was pumped from a constant temperature tank, while the hot liquid from the column was returned to a separate tank. Following a set of runs the hot liquid was circulated through the heat exchanger till it was at the proper temperature for the next set of runs.

Packing properties were listed in an earlier paper (*10*) and elsewhere (*2*). Thermal conductivities of the packing material ranged from 0.1 to 0.3 B.t.u./(hr.) (ft.) (°F.).

HEAT TRANSFER

If axial conduction is considered negligible and rodlike flow of both gas and liquid is assumed, then the partial differential equation for a cylindrical section of packed column in concurrent gas liquid flow with equal gas, liquid, and solid temperature will be

$$(G_g C p_g{}^* + G_l C p_l) \frac{\partial T}{\partial z} = K_e \left(\frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial r^2} \right) \tag{1}$$

All resistance to heat transfer is included in this effective thermal conductivity. These resistances include thermal resistance at the wall, thermal resistance of the particles and of the contact area between the particles, thermal resistance of the liquid and gas fillets between particles, thermal resistance from the particles to the liquid, thermal resistance from the liquid surrounding particle to the bulk of the gas, and thermal resistance of both the liquid and the gas at rest and in motion.

The general solution of Equation (1) in terms of the dimensionless temperature Δ can be shown to be (*3*)

$$\Delta = C_1 e^{-Ha^2 z} \left[C_2 J_0(ar) + C_3 Y_0(ar) \right] \tag{2}$$

The necessary boundary conditions are

when $z = 0, \quad \Delta = 1$
when $z = \infty, \quad \Delta = 0$
when $r = 0, \quad \partial\Delta/\partial r = 0$
when $r = 0, \quad \Delta = \text{finite}$
when $r = r_0, \quad \Delta = 0$

Application of these boundary conditions leads to a series solution in terms of the bulk outlet temperature:

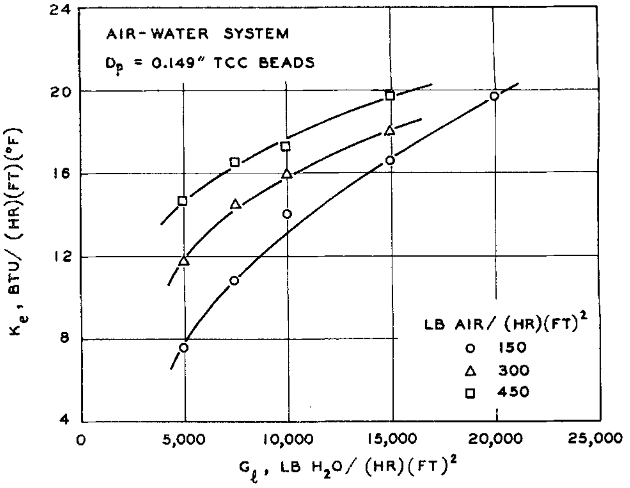

Fig. 2. Effect of liquid rate on effective thermal conductivity.

$$\frac{T_w - T_b}{T_w - T_0} = 4 \sum_{n=1}^{\infty} \frac{1}{(\delta_n)^2} e^{-H\delta_n{}^2 \, z/r_0{}^2} \tag{3}$$

Newman (*4*) evaluated this series, and his tabulated results were used to calculate the K_e. Knowing only the inlet temperature, average wall temperature, and the bulk outlet temperature one can solve for K_e from Equation (3).

An equivalent heat transfer coefficient h_o may be defined as follows:

$$h_o = \frac{Q}{2\pi r_o z \Delta T_{lm}} \tag{4}$$

Since Equation (3) represents a rapidly converging series, it is possible to neglect all but the first term as a good approximation. Thus, when T_b is defined from the first term of Equation (3) and substituted into Equation (4), the following expression results:

$$h_o = \frac{2.892}{r_o} K_e + \frac{0.183 r_o \, (G_l C p_l + G_g C p_g{}^*)}{z} \tag{5}$$

Where vaporization may take place, as in the air-water system, the correct heat balance can be achieved using $Cp_g{}^* = \Delta H_g / \Delta T_g$ instead of Cp_g. Over relatively small temperature ranges (that is from 70° to 150°F.) the $Cp_g{}^*$ may be treated as a constant. Wet and dry bulb readings of the inlet and outlet air to the heat transfer section showed that even at the highest gas rates and lowest liquid rates the air was saturated.

As might be anticipated, the transition from gas-continuous to pulse flow had an important effect on the heat transfer. Figure 1 shows the effect of transition on K_e at three liquid rates for the glass beads. It can be seen that significant increases in both K_e and h_o occur at gas rates which correspond closely to the transition gas rates given in a previous article (*10*). Similar increases in heat transfer were also observed for the other two packing sizes.

In general, both K_e and h_o increased with increasing gas and liquid rates. Figure 2 shows the effect of increasing liquid rate on K_e for various gas rates.

RADIAL TRANSPORT MODEL

A simple model for radial heat transfer can be constructed by assuming that all the various heat transfer mechanisms can be lumped into two terms (*5*). All heat transferred by eddy diffusion in the radial direction would constitute one term, while the molecular conduction in the stagnant bed would represent the other.

Thus, the expression for the effective conductivity in terms of the stagnant contribution $(K_e)_o$ and the radial mixing contribution $(K_e)_t$ would be

$$K_e = (K_e)_o + (K_e)_t \tag{6}$$

The radial mixing contribution may be further separated into that due to the gas and that due to the liquid:

$$(K_e)_t = (K_e)_{tl} + (K_e)_{tg} \tag{7}$$

It is possible to relate the heat being transferred radially owing to $(K_e)_t$ to that carried by the lateral or radial component of the flow. The fraction of the mass velocity having a radial direction will be defined as γ.

At a given radial position r, the heat flux may be expressed as the product of $(K_e)_{tl}$ and the radial temperature gradient. The flux may also be written in terms of heat transmitted by the radial motion of the fluid as follows:

$$Q/A = (K_e)_{tl} \left(\frac{dT}{dr}\right) = \gamma U_o \rho_l Cp_l (T_w - T) \tag{8}$$

Recalling that T is given as a function of r by Equation (2) and utilizing only the first term of the series, one may rewrite Equation (8) as

$$\frac{(K_e)_{tl}}{\gamma} = U_o Cp_l \rho_l \left[\frac{r_o}{\delta_l} \frac{J_o(\delta_1 r/r_o)}{J_1(\delta_1 r/r_o)}\right] = U_o Cp_l \rho_l \varphi(r) \tag{9}$$

It can be seen from Equation (9) that if $(K_e)_{tl}$ is assumed constant, then γ will be some function of r_o.

Ranz (6) defined γ for tetrahedral stacking of spheres and assumed the $\varphi(r)$ was equal to the diameter of the packing. In the present work it was observed that the amount of heat transferred did not appear to be a function of the packing size. It therefore seemed reasonable to define the $\varphi(r)$ as equal to the tube diameter (D_t). The expression for $(K_e)_{tl}$ then becomes

$$(K_e)_{tl} = \gamma U_o \rho_l Cp_l D_t \tag{10}$$

Golpalarathnam, Hoelscher, and Laddha (7) (hereafter referred to as G.H.L.) also defined $(K_e)_{tl}$ for single-phase liquid flow and defined the $\varphi(r)$ as D_t. The effective thermal conductivity may now be written as

$$K_e = (K_e)_o + \gamma G_l Cp_l D_t + \beta G_g Cp_g D_t \tag{11}$$

Dividing Equation (11) by k_l and multiplying the last two terms by μ_l/μ_l and $\mu_g k_g/\mu_g k_g$, respectively, one obtains the following after rewriting in terms of Reynolds and Prandtl numbers:

$$\frac{K_e}{k_l} = \frac{(K_e)_o}{k_l} + \gamma (N_{Re})_l (N_{Pr})_l + \beta \left(\frac{k_g}{k_l}\right) (N_{Re})_g (N_{Pr})_g \tag{12}$$

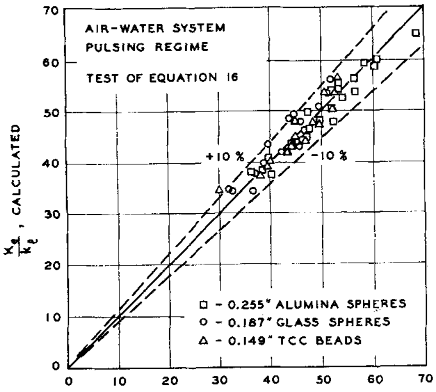

Fig. 3. Test of radial transport correlation.

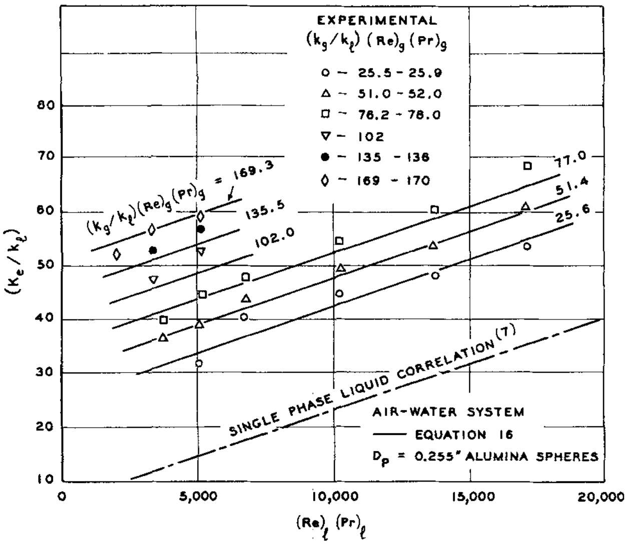

Fig. 4. Comparison of radial transport equation with experimental data.

With the values of K_e obtained in the pulsing region for all three packing sizes, the constants $(K_e)_o$, γ, and β were determined by the method of least squares. The numerical values obtained were

$$(K_e)_o = 7.71$$
$$\gamma = 0.00174$$
$$\beta = 0.172$$

Figure 3 shows the experimentally determined K_e/k_l values and those calculated by Equation (12). The maximum deviation was 10%, and the average deviation was approximately 5%.

Figure 4 compares experimental data for the 0.255-in. alumina spheres to the correlation obtained from Equation (12) and also shows the equation of G.H.L. (7) for single-phase liquid flow. It can be seen that at constant gas rate, Equation (12) gives essentially the same slope as that given by G.H.L. for single-phase liquid flow. At constant gas rate, γ is 0.00174, while G.H.L. found the slope to be 0.00176. This remarkably close agreement implies that for both single-phase and two-phase flow, the fraction of the total liquid mass velocity having a radial component is the same. The primary effect of the gas is to impart a greater velocity to the liquid phase. Thus, while the fraction of the liquid velocity in the radial direction is the same, the radial component will be larger for two-phase flow. A substantial fraction of the total radial transport of heat is probably accomplished by the liquid pulses which occur naturally in this system. It is likely that within these liquid pulses the ratio of the liquid having a radial component to the total liquid flow is the same as with single-phase flow. Figure 4 shows that the presence of the air gives a substantial increase in the effective thermal conductivity ranging from 200 to 400%.

By employing the Larkin et al. (1) correlation for the liquid saturation, it is possible to calculate the liquid mass velocity based only on the liquid flow area and thus account for the increased liquid velocity due to the presence of the air. The liquid mass velocity based on the actual

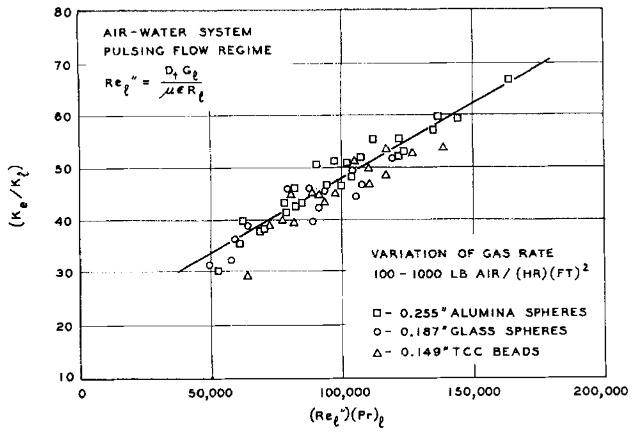

Fig. 5. Radial transport correlation liquid-phase basis.

area available for liquid flow is

$$G_l' = \frac{W_l}{A_o \epsilon R_l} = \frac{G_l}{\epsilon R_l} \tag{13}$$

Thus, the Reynolds number becomes

$$(N''_{Re})_l = \frac{D_t G_l}{\mu_l \epsilon R_l} \tag{14}$$

The effect of gas rate on the liquid velocity can now be accounted for, since as the gas rate increases, R_l decreases and consequently $(N''_{Re})_l$ increases. Figure 5 shows that this definition of $(N''_{Re})_l$ gives a single correlation for all gas rates and satisfactorily accounts for the increase in the radial liquid velocity due to the presence of the gas phase. The final form of this correlation was determined by the method of least squares to be

$$\frac{K_e}{k_l} = \frac{7.03}{k_l} + 0.000285\,(N''_{Re})_l\,(N_{Pr})_l \tag{15}$$

PENETRATION MODEL

The penetration theory (8) has often proved to be an effective tool in analyzing heat and mass transfer. In essence, the theory presumes a short period of unsteady state heating, after which complete mixing occurs and another unsteady state period begins. An important restraint on the theory is the necessity of keeping the contact time (that is unsteady heating period) as short as possible. This restraint is imposed since the theory assumes that the temperature or concentration profiles develop undisturbed during the unsteady state period. With relatively long contact times, these profiles will penetrate far enough from the wall that they will be disturbed by the main flow.

To apply penetration theory to a two-phase gas-liquid system in a packed bed, it is necessary to assume that during the contact time there is a quiescent layer of liquid at the wall. Visual observations with the glass section have indicated that this layer is indeed present at the wall between pulses. Thus, the unsteady state temperature profiles will build up in the liquid layer near the wall; the gas phase affects only the contact time.

Because of the restraint of short contact times and the resultant short penetration distances involved, negligible error is introduced by writing the defining equation for a planar wall as opposed to a cylindrical wall. The unsteady state equation for conduction in the liquid layer close to the wall will be

$$\frac{\partial \Delta}{\partial \theta} = \alpha_l \frac{\partial^2 \Delta}{\partial y^2} \tag{16}$$

The necessary boundary conditions are

$$\begin{aligned} \Delta &= 1, \quad y \geqq 0, \quad \theta = 0 \\ \Delta &= 0, \quad y = 0, \quad \theta > 0 \\ \Delta &= 1, \quad y = \infty, \quad \theta \geqq 0 \end{aligned}$$

The solution can be expressed in terms of the error function (erf) as follows (9):

$$\Delta = \text{erf}\,\frac{y}{2\sqrt{\alpha_l \theta}} \tag{17}$$

Based on the third boundary condition, the equivalent heat transfer may be written as

$$h_o = \frac{Q/A}{T_w - T_o} \tag{18}$$

Based on contact time θ_c, the equivalent heat transfer coefficient h_o can be shown to be (9)

$$h_o = 2\rho_l\, Cp_l \sqrt{\frac{\alpha_l}{\pi \theta_c}} \tag{19}$$

The major problem in applying penetration theory is properly defining the contact time θ_c. The presence of the pulses in the two-phase system provided a convenient basis for measuring the contact time. If it is assumed that the pulses completely mix the fluid at the wall and sweep away the temperature profiles established during the heating period, and further that the pulses have a negligible vertical thickness, then the contact time θ_c will simply be the reciprocal of the pulse frequency. Thus

$$\theta_c = 1/F_p$$

Equation (19) now becomes

$$h_o = 2\rho_l\, Cp_l \sqrt{\frac{\alpha_l F_p}{\pi}} \tag{20}$$

It should be noted that this equation has been derived entirely on a theoretical basis and does not contain any experimental constants.

The experimentally determined equivalent heat transfer coefficient h_o has been plotted vs. F_p in Figure 6. Shown for comparison on this figure is Equation (20) plotted with the average value of the thermal diffusivity (α_l = 1.64 × 10^{-6} sq.ft./sec.) for range of data given. It can be seen that the penetration theory predicts the heat transfer data remarkably well, considering the scope of the assumptions made. The penetration theory not only predicts

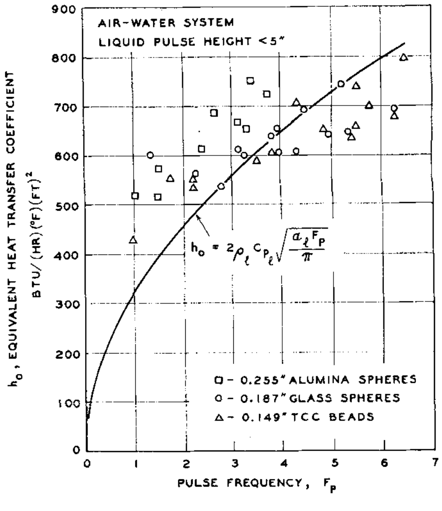

Fig. 6. Comparison of penetration theory with experimental data.

the order of magnitude but differs by only ± 20% from the bulk of the data.

The greatest deviations from Equation (20) were observed where the liquid pulse height h_l was large in comparison with the height of a pulsing unit H_t and where contact time was greater than 0.5 sec. (that is $F_p < 2$). These deviations are to be expected, since these are the conditions which are farthest from the assumptions made in deriving Equation (20). As the liquid pulses become longer, the unsteady heating period becomes shorter (that is θ_c becomes smaller) even though the frequency F_p is decreasing. There is, of course, some heat being transferred into the liquid pulses which is not accounted for by Equation (20). At the other extreme, where the liquid pulse height h_l is very small (that is <1 in.) and the frequency low, the contact time is large enough to allow the unsteady state temperature profiles to penetrate into the turbulent region and thus be disturbed.

SUMMARY

The transition from gas-continuous to pulsing flow results in a large increase (certain cases > 400%) in the radial heat transfer in the two-phase system. The presence of the flowing gas phase greatly increases the effective thermal conductivity over that observed for single-phase liquid flow. Lumped heat transfer coefficients at the wall were remarkably high, with a value of $h = 1{,}100$ B.t.u./(hr.) (°F.) (sq.ft.) observed at $G_l = 25{,}000$ lb. water/(hr.) (sq.ft.) and $G_g = 450$ lb. air/(hr.) (sq.ft.).

It was possible to obtain a good correlation of the experimental data based on a simple radial transport model. The heat transfer mechanism in the pulsing regime may be explained by application of penetration theory.

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NOTATION

a = constant determined by boundary conditions
A_o = cross-sectional area of empty tube, sq.ft.
A_g = cross-sectional area for gas flow, sq.ft.
A_l = cross-sectional area for liquid flow, sq.ft.
Cp = specific heat, B.t.u./(lb.) (°F.)
Cp_g^* = specific heat of saturated air, $\Delta H_g/\Delta T_g$, B.t.u./(lb.) (°F.)
D = tube diameter, ft.
D_p = particle diameter, ft.
F_p = pulse frequency, pulses/sec.
G = mass velocity based on empty tube area, lb./(hr.) (sq.ft.)
H = $K_e/(G_lCp_l + G_gCp_g^*)$
h = height of liquid or gas section of pulse, ft.
h_o = equivalent heat transfer coefficient, B.t.u./(hr.) (sq.ft.) (°F.)
H_t = total height of a pulsing unit, ft.
H_g = enthalpy of saturated air, B.t.u./lb.
J_o, Y_o = zero-order Bessel functions of first and second kind, respectively
k = molecular conductivity of liquid or gas, B.t.u./(hr.) (ft.) (°F.)
K_e = overall effective thermal conductivity, B.t.u./(hr.) (ft.) (°F.)
$(K_e)_t$ = contribution to overall effective thermal conductivity due to turbulent radial movement of liquid or gas, B.t.u./(hr.) (ft.) (°F.)
$(K_e)_o$ = contribution to overall effective thermal conductivity due to molecular conduction in stagnant bed, B.t.u./(hr.) (ft.) (°F.)
N_{Pr} = Prandtl number, $Cp\ \mu/k$
N_{Re} = Reynolds number, $\frac{D_tG}{\mu}$
N_{Re}'' = modified Reynolds number, $\frac{D_tG}{\mu\epsilon R}$
Q = heat transfer rate, B.t.u./(hr.)
R = fraction of void volume occupied by gas or liquid
r = radius at any point, ft.
r_o = radius of tube, ft.
T = temperature at r, °F.
T_b = outlet mixing cup temperature, °F.
T_o = inlet bulk temperature, °F.
T_w = wall temperature, °F.
U_o = superficial velocity based on empty tube, ft./hr.
W = mass rate, lb./(hr.)
y = distance from wall, ft.
z = axial distance, ft.

Greek Letters

α = thermal diffusivity, $k/\rho Cp$
γ, β = radial fraction of liquid and gas mass velocity
δ = zero roots of first order Bessel function J_o
Δ = dimensionless temperature, $(T_w - T)/(T_w - T_o)$
ΔH_g = enthalpy increase of moist air
ΔT_{lm} = log mean temperature difference, $\left[(T_w - T_o) - (T_w - T_b)\right] \Big/ \ln \frac{T_w - T_o}{T_w - T_b}$
ΔT_g = $T_o - T_b$
ϵ = fraction void space in packed bed
θ = time, sec.
θ_c = contact time, sec.
μ = viscosity, lb./(hr.) (ft.)
ρ = density, lb./cu.ft.

Subscripts

g = gas
l = liquid
p = pulse
t = turbulent
tg = turbulent gas
tl = turbulent liquid

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