accuracy of the Klee and Treybal terminal velocity correlation (± 4.5%), the precision of the statistically determined Sauter mean diameter (±8%), and the errors inherent in the Zenz correlation itself. It is possible that the use of an alternate correlation rather than that of Zenz for predicting the holdup from the analogue solid-fluidized system might increase the agreement between the experimental and predicted curves. Such an alternate correlation has been given by Richardson and Zaki (*6*).

A curve relating holdup to slip velocity can be constructed readily from the above predicted curve for the case of a spray column operating with a quiescent continuous phase. But, as demonstrated earlier, the relationship of holdup to slip velocity in a nonflooded spray column is independent of continuous-phase flow. Thus the holdup vs. slip-velocity curve for a nonflooded spray column with a quiescent or flowing continuous phase can be predicted from the solid-in-liquid analogue system.

NOTATION

V_s = slip velocity
V_t = single droplet terminal velocity
Q_d = volumetric flow rate of disperse phase
Q_c = volumetric flow rate of continuous phase
A = column cross-sectional area
ϵ = fraction void or fraction of column occupied by the continuous phase
$1-\epsilon$ = fraction holdup of disperse phase

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Fluid - Particle Mass Transfer in a Packed Bed

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Mass transfer coefficients have been measured for air flowing through a bed of naphthalene pellets. Results were obtained for 3/8-in. spheres and for 1/2, 3/8, and 1/4-in. cylinders in a bed 4 in. in diameter and packed to heights varying from 5 to 10 in. Radial concentration profiles were obtained for some conditions, from which the radial variation of the mass transfer coefficient was determined. The point-values of k_g follow an equation of the form $k_g = a\,G^b$. From mixed outlet concentrations, values of $k_{g_{av}}$ and j_d, corrected for the effect of axial diffusion, have been obtained for all the pellet sizes as functions of mass velocity and Reynolds number, respectively.

Since it is known that the velocity in a packed bed varies with the radial position (*31*), it is of interest to determine how the mass transfer coefficient varies with radial position. The coefficient should depend on the velocity in a systematic way, and the results of the present work show that it does. Another investigator (*33*) found on the contrary that k_g was not a function of radial position. Local coefficients in the present work were measured at room temperature for a bed of 1/2-in. cylinders of naphthalene evaporating into air for a range of $D_p\, G_{av}/\mu$ from 840 to 9,900 for bed heights of about 4.5 and 6.0 in.

Although many studies of mass transfer in packed beds have been made (*1, 4, 10, 11, 14, 16, 17, 18, 19, 20, 21, 22, 25, 27, 28, 29, 30, 32, 33, 34, 36, 37*), apparently none have been made in which the effect of axial dispersion has been evaluated quantitatively. As a matter of fact Ergun (*13*) has asserted that many earlier studies on solid-gas systems actually were made on beds in which there was complete mixing. Therefore in the present study not only has the configuration of the bed been designed to reduce the importance of axial eddy diffusion, but the mass transfer coefficients have also been corrected for the

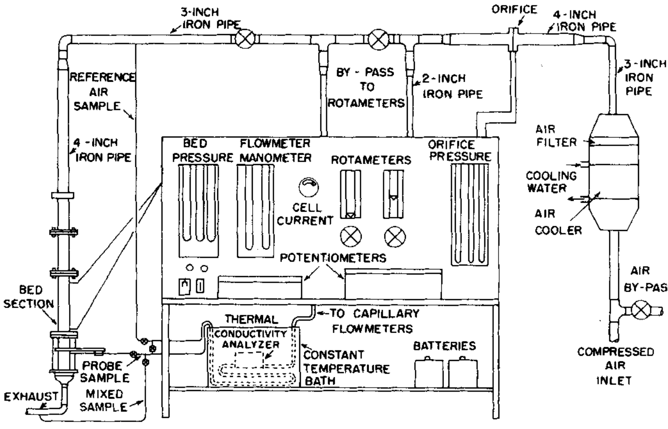

Fig. 1. Diagram of experimental apparatus.

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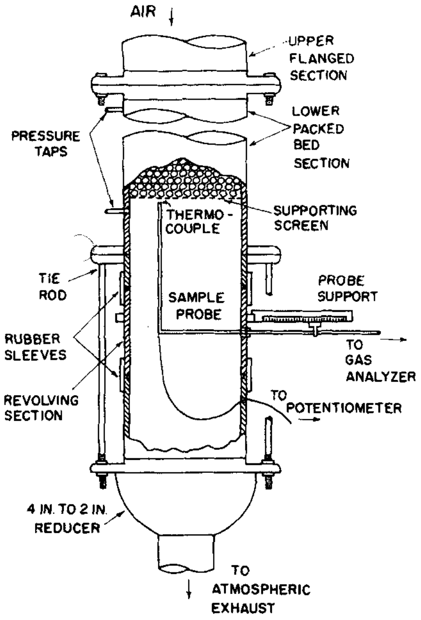

Fig. 2. Detail of bed and sampling sections.

amount of axial diffusion actually present. Over the past few years several publications have appeared which make the latter calculation possible (*3, 8, 12, 23, 35*). In the present work average coefficients corrected for axial diffusion effects have been measured at room temperature for ⅜-in. spheres and ½, ⅜, and ¼-in. cylinders of naphthalene evaporating into air at Reynolds numbers ranging from 438 to 9,900.

EXPERIMENTAL METHOD

The general appearance of the apparatus used in this work is shown in Figure 1; the blower, designed to deliver 300 s.c.f.m. at 15 lb./sq. in. gauge is not shown. The packed-bed section is shown in Figure 2. The bed was supported by an 8-mesh screen; the air flowed downward. The figure shows a section of pipe below the bed which could be revolved; this arrangement made it possible to position the probe at any point in the pipe cross section. The naphthalene concentrations were measured by a thermal conductivity cell; details of its use and calibration are available (*7*). The pellets were punched from flake naphthalene with tablet machines.

METHOD OF CALCULATION OF POINT COEFFICIENTS

The value of the mass transfer coefficient at various points within a packed bed can be computed from the observed data by the use of a differential mass balance written for the naphthalene. Starting from the basic analysis of Bird, Stewart, and Lightfoot (*5*) one can write the differential balance in cylindrical coordinates as

$$u_r \frac{\partial c}{\partial r} + u_\theta \frac{1}{r}\frac{\partial c}{\partial \theta} + u_z \frac{\partial c}{\partial z} + \frac{\partial c}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\,E'_r \frac{\partial c}{\partial r}\right) + \frac{1}{r}\frac{\partial}{\partial \theta}\left(\frac{1}{r}E'_\theta \frac{\partial c}{\partial \theta}\right) + \frac{\partial}{\partial z}\left(E'_z \frac{\partial c}{\partial z}\right) + \frac{6(1-\epsilon)k_c(c_s - c)(1-y)}{D_p} \qquad (1)$$

Since the flow of air is steady and axially symmetric, the left-hand side of this equation reduces to $u_z(\partial c/\partial z)$, and the second term on the right is zero.

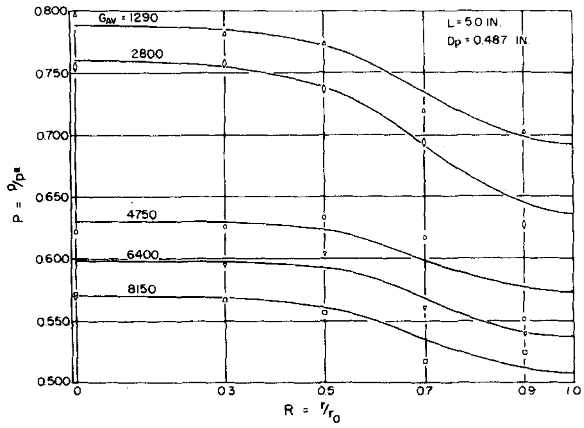

Fig. 3. Radial concentration profiles.

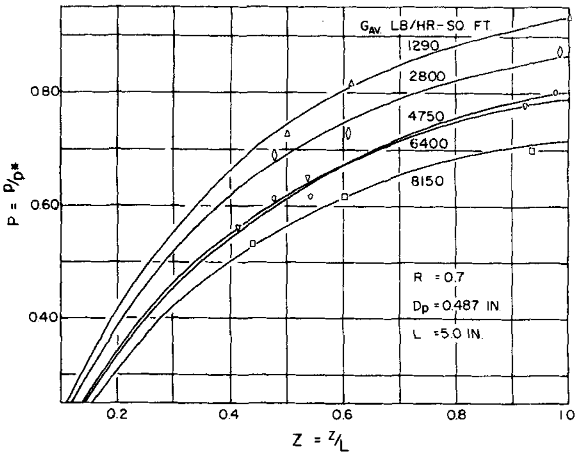

Fig. 4. Axial concentration profiles.

The mass balance has been made on a differential element of the packed bed, and u_z is thus the superficial velocity at a point. This fact is emphasized by replacing the u_z by u_s in the equations to follow. The superficial velocity is related to the interstitial velocity by

$$u_s = u\,\epsilon \qquad (2)$$

E'_r, E'_θ, and E'_z are based on the total area perpendicular to the direction in question. The following relations are usually considered to hold:

$$E' = E\,\epsilon \qquad (3)$$

$$E = E_t + D \qquad (4)$$

$$N_{Pe} = \frac{D_p u}{E} = \frac{D_p u_s}{E'} \qquad (5)$$

It should be noted that Equations (2) and (3) are only approximations; the tortuous nature of the paths between the particles should also be considered. This point has been investigated by Carman (*9*) with regard to Equation (2). It is well known that the effective molecular diffusivity is not equal to $D\,\epsilon$; refer for example to the work of Bokhoven and van Raayen (*6*). However these points are of no practical importance here, since experimental measurements of the Peclet number are based on the measurements of E' and u_s, even though the results may be given as $D_p\, u/E$.

The dimensions of the pellets before and after runs in this work differ by several per cent; details of the calculation of an average D_p are given in reference *7*. For naphthalene in air y is so small that $1 - y \cong 1$. Equation (1) is now rewritten, incorporating the simplifications listed in the intervening discussion and using dimensionless variables:

$$\frac{\partial P}{\partial Z} = \left(\frac{L\,E'_r}{u_s r_o^2}\right)\frac{1}{R}\frac{\partial P}{\partial R} + \frac{L}{u_s r_o^2}\left(\frac{\partial E'_r}{\partial R}\right)\frac{\partial P}{\partial R} + \left(\frac{L\,E'_r}{u_s r_o^2}\right)\frac{\partial^2 P}{\partial R^2}$$

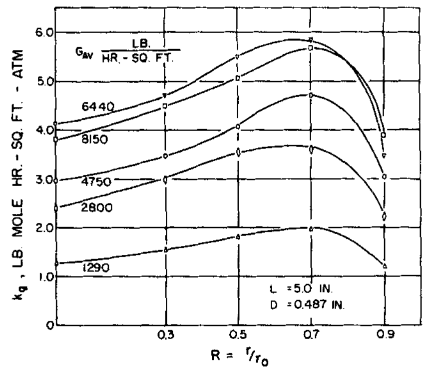

Fig. 5. Values of the mass transfer coefficient as a function of radial position.

Table 1. Calculation of Point Values of k_g

R	E'_r	$u_s/u_{s_{av}}$	ϵ	$1-P$	$\partial P/\partial Z$	$\frac{LE'_r}{u_s r_o^2}\frac{1}{R}\frac{\partial P}{\partial R}$	$\frac{L}{u_s r_o^2}\frac{\partial E'_r}{\partial R}\frac{\partial P}{\partial R}$	$\frac{LE'_r}{u_s r_o^2}\frac{\partial^2 P}{\partial R^2}$	$\frac{D_p}{N_{Pe_z} L}\frac{\partial^2 P}{\partial Z^2}$	k_g
0	0.0606	0.70	0.36	0.379	0.741	0	0	0	−0.0408	2.98
0.3	0.0617	0.74	0.42	0.390	0.751	−0.0019	−0.0001	−0.0062	−0.0430	3.46
0.5	0.0635	0.83	0.44	0.394	0.716	−0.0118	+0.0001	−0.0462	−0.0416	4.05
0.7	0.0550	0.98	0.52	0.413	0.705	−0.0216	+0.0244	+0.0146	−0.0334	4.70
0.9	0.0348	1.07	0.27	0.451	0.715	−0.0052	+0.0172	+0.0338	−0.0372	3.03

$u_{s_{av}} = 17.35$ $D_p = 0.0402$ $RT = 387.6$ $z = 0.398$

$G_{av} = 4746$ $r_o = 0.1677$ $N_{Pe_z} = 2.0$ $L = 0.815$, deepest bed

$$+\left(\frac{D_p}{N_{Pe_z}L}\right)\frac{\partial^2 P}{\partial Z^2} + \frac{6(1-\epsilon)RT\,k_g(1-P)L}{D_p u_s} \qquad (6)$$

It is necessary to retain the term for radial diffusion because the velocity profile in the bed (*31*) causes the rate of generation of naphthalene to vary with radial position in the bed. The data of Fahien and Smith (*15*) show that E'_r is also a function of r, so it must be left inside the parenthesis in Equation (6). On the other hand E'_z apparently does not vary with z. As a matter of fact for the high Reynolds numbers used in this work N_{Pe_z} is taken as a constant, 2.0 (*23*).

RESULTS FOR POINT COEFFICIENTS

Some typical results for the radial concentration profile with the ½-in. cylinders are given in Figure 3. Each point is the average of four determinations. Similar curves were obtained for other bed heights (*7*). The curves in Figure 3 were not drawn to pass through all the points but were drawn so that $\partial P/\partial R$ was zero at $R = 0$ and at $R = 1.0$. Plots were also made of $\partial P/\partial R$ vs. R (*7*).

Figure 4 shows some typical axial concentration profiles; similar curves were obtained at other radii, as well as curves of $\partial P/\partial Z$ vs. Z (*7*). In obtaining these results it has been assumed that the concentrations at the outlet of a bed z_1 feet high are the same as those which would exist at corresponding positions inside a longer bed, z_1 feet from the bed entrance.

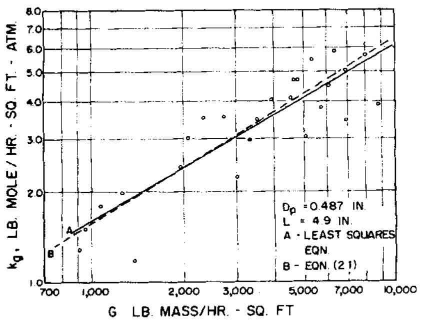

Fig. 6. Correlation between the point values of k_g and G.

By the graphical differentiation of the various concentration profiles and by the use of data on the radial variation of E'_r (*15*), u_s (*31*), and ϵ (*26*), values of k_g were calculated by using Equation (6). The results, which are independent of bed height, are shown in Figure 5. The magnitudes of the individual terms in Equation (6) are illustrated by the information in Table 1.

In view of the method by which the axial concentration profile was constructed from the outlet concentrations of beds of varying length it is questionable whether to include the term for axial diffusion. The axial term has been included, but it is of minor interest in the present calculation, which is primarily aimed at showing the radial variation in k_g.

Figure 6 is a plot of the values of the point mass transfer coefficients vs. the superficial point mass velocity. The following equation, obtained by a least-squares analysis, fits the data with a standard deviation of 24.9%:

$$k_g = 0.0296\,G^{0.577} \qquad (7)$$

This standard deviation is only moderately higher than that obtained from a similar representation of the average coefficients, to be given presently. This similarity is further shown by a plot of Equation (21) obtained for the average coefficients, shown on Figure 6 by a dotted line.

METHOD OF CALCULATION OF AVERAGE COEFFICIENTS

Theoretically Equation (7) and analytical expressions for u_s, ϵ, and E'_r could be used to integrate Equation (6), but the mathematical difficulty of this approach is excessive. If a chemical reaction is taking place in the bed and heat flows through the tube walls, the radial heat and mass transfer effects cannot be neglected. This type of problem has been solved for constant k_g, u_s, ϵ, and E'_r by Amundsen (*2*). However if these four quantities are assumed constant for the packed bed used in the present work, the radial concentration gradients disappear, for they are caused by the radial variation of the generation term, which in turn is caused by the radial variation of u_s and ϵ. When one omits the radial terms, Equation (6) becomes

$$\frac{d^2P}{dZ^2} - \frac{N_{Pe_z}L}{D_p}\frac{dP}{dZ} + \frac{6(1-\epsilon)_{av}RT\,k_{g_{av}}N_{Pe_z}L^2(1-P)}{D_p^2\,u_{s_{av}}} = 0 \qquad (8)$$

It is convenient to define a new variable f equal to $1-P$ and write

$$\frac{1}{N_{Pe_L}}\frac{d^2f}{dZ^2} - \frac{df}{dZ} - R_1 f = 0 \qquad (9)$$

in which

$$N_{Pe_L} = \frac{N_{Pe_z}L}{D_p} \cong \frac{2L}{D_p} \qquad (10)$$

and

$$R_1 = \frac{6(1-\epsilon)_{av}RT\,k_{g_{av}}L}{D_p\,u_{s_{av}}} \qquad (11)$$

The boundary conditions to use with Equation (9) are by no means obvious. In accordance with the careful analysis of Wehner and Wilhelm (*35*)

$$f - \frac{1}{N_{Pe_L}}\frac{df}{dZ} = 1.0;\ Z = 0 \qquad (12)$$

$$\frac{df}{dZ} = 0;\ Z = 1.0 \qquad (13)$$

The solution (*35*) of Equation (9) at $Z = 1.0$ is then

$$f_1 = \frac{4a\exp\left(\frac{N_{Pe_L}}{2}\right)}{(1+a)^2\exp\left(\frac{aN_{Pe_L}}{2}\right) - (1-a)^2\exp\left(\frac{-aN_{Pe_L}}{2}\right)} \qquad (14)$$

in which

$$a = \left(1 + \frac{4R_1}{N_{Pe_L}}\right)^{1/2} \qquad (15)$$

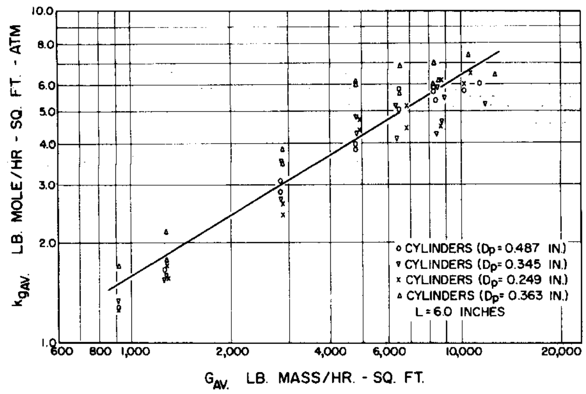

Fig. 7. Correlation between the values of $k_{g_{av}}$ and G_{av}.

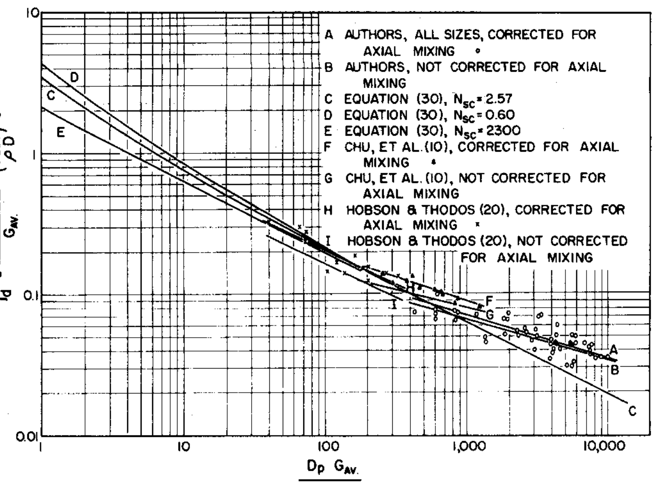

Fig. 8. Mass transfer factor for packed beds.

In Equation (14) f_1 is the average concentration at $Z = 1.0$; this quantity corresponds to a concentration measured at some distance downstream from the bed outlet. At this point the radial gradients of concentration have disappeared.

The boundary conditions represented by Equations (12) and (13) are based on the assumption of a continuous variation in concentration in the bed and in the sections upstream and downstream from the bed. The bed may alternatively be viewed as a series of void spaces of length approximately D_p; these spaces act like mixing tanks (*3*). The number of perfect mixing tanks is given by

$$n = \frac{Lu_s}{2\,E'_z} = \frac{L\,N_{Pe_z}}{D_p\,2} \qquad (16)$$

At Reynolds numbers below approximately 1,000, N_{Pe_z} is less than 2; this condition probably corresponds to some bypassing or channeling, so $n < L/D_p$ (*8*). For highly turbulent flow N_{Pe_z} approaches 2.0, so that the number of equivalent perfect mixers is equal to L/D_p. This model calls for finite discontinuities in concentration in passing from the section before the bed to the first mixer and from one mixer to the next.

Epstein (*12*) has shown that the mixer model leads to the following equations for the present situation:

$$R_1 = n\left[\left(\frac{1}{f_1}\right)^{\frac{1}{n}} - 1\right] \qquad (17)$$

and solving for f_1 one obtains

$$f_1 = \left(\frac{n}{R_1 + n}\right)^n \qquad (18)$$

For a bed with complete mixing n equals unity, and Equation (18) must of course reduce to an equation of the type used by Ergun (*13*) in his analysis:

$$f_1 = \frac{1}{R_1 + 1} \qquad (19)$$

If there is no axial mixing, n is infinite and equation (18) must become

$$f_1 = \exp\,(-R_1) \qquad (20)$$

As n becomes large, the differences between the analysis based on a continuous and a step-by-step concentration variation disappears (*3, 8*). Experimental studies on axial diffusion have been made in beds having a large n (*8, 23*). The value of n (that is L/D_p) in the present work is as low as 10 and has been even lower in many previous investigations; therefore Equation (17) is used in preference to Equation (14). The boundary conditions, Equations (12) and (13), are not valid for small values of n, such as those used in this work.

RESULTS FOR AVERAGE COEFFICIENTS

The results of all the measurements of $k_{g_{av}}$ calculated by the use of Equation (17) are shown in Figure 7. No statistically significant effect of D_p/D_t was discerned. The equation

$$k_{g_{av}} = 0.0219 G_{av}^{0.617} \qquad (21)$$

fits the data with a standard deviation of 17.8%. Table 2 shows the importance of the correction for axial diffusion for this work and for some of the few cases from the literature for which enough data were given to calculate the effect. As suggested by Ergun (*13*) axial mixing is often extremely important. The ratios $k_{g_{av}}/k'_{g_{av}}$ have been calculated from Equations (17) and (20), with n equal to L/D_p.

It is usual to correlate mass transfer rates in a packed bed by using the factor j_d defined by

$$j_d = \frac{k_{g_{av}}\,R\,T\,\rho}{G_{av}}\left(\frac{\mu}{\rho D}\right)^{2/3} \qquad (22)$$

This quantity is plotted against $D_p\,G_{av}/\mu$ in Figure 8; the equation for a straight line (A) through the points is given by

$$j_d = 0.606\left(\frac{D_p\,G_{av}}{\mu}\right)^{-0.309} \qquad (23)$$

with $\sigma = 18.7\%$. Also shown is a line (B) representing j_d calculated from $k_{g_{av}}$ by omitting the axial mixing effect [Equation (20)]; its equation is

$$j_d = 0.506\left(\frac{D_p\,G_{av}}{\mu}\right)^{-0.293} \qquad (24)$$

Table 2. Effect of Axial Diffusion on $k_{g_{av}}$

Investigator	D_p	D_t	L	G_{av}	f_1	$\frac{k_{g_{av}}}{k'_{g_{av}}}$
Authors						
3-6	0.0197	0.334	0.450	8,660	0.222	1.038
2-4	0.0285	0.334	0.505	4,850	0.194	1.044
4-1	0.0302	0.334	0.510	919	0.070	1.086
Chu, et al. (*10*)						
108	0.0443	0.312	0.167	1,200	0.500	1.100
68	0.0282	0.312	0.083	1,040	0.490	1.132
61	0.0173	0.312	0.041	470	0.378	1.236
Hobson and Thodos (*20*)						
BAI	0.0308	0.172	0.087	150	0.470	1.147
OA2	0.0308	0.172	0.092	55	0.241	1.281
WA4	0.0308	0.172	0.126	387	0.128	1.303

with $\sigma = 18.0\%$. The use of $D_p G_{av}/(1-e)\mu$ as the abscissa, as suggested by Ergun (*13*), did not reduce the standard deviations.

It is interesting to attempt to relate the data for the packed beds to the data for mass transfer from a single sphere by using the approach suggested by Ranz (*24*). For a single sphere the equation of Froessling is valid up to N_{Re} of about 10,000:

$$\frac{k_{g_{av}} RTD_p}{D} = 2.0 + 0.60 \left(\frac{\mu}{\rho D}\right)^{1/3} \left(\frac{D_p G_{av}}{\mu}\right)^{1/2} \qquad (25)$$

This equation can be combined with the definition of j_d to yield

$$j_{d_s} = \frac{2.0}{N_{Re} N_{Sc}^{1/3}} + \frac{0.60}{N_{Re}^{1/2}} \qquad (26)$$

In accordance with Ranz (*24*) for a packed bed the mass velocity is multiplied by 10.73, and j_d for this configuration is then

$$j_d = \frac{2.0}{N_{Re} N_{Sc}^{1/3}} + \frac{1.97}{N_{Re}^{1/2}} \qquad (27)$$

Equation (27), with $N_{Sc} = 2.57$ for the naphthalene—air system, is plotted (C) in Figure 8. Lines are also plotted for $N_{Sc} = 0.60$ (D) (water-air) and for $N_{Sc} = 2{,}300$ (E) (benzoic acid-water).

The omission of the correction for axial diffusion changes not only the magnitude of j_d but also the exponent in an expression like Equation (23). At low Reynolds number the effect of molecular diffusion becomes increasingly important, and this effect may also cause deviations from an equation like (23).

The pressure drop across the packed bed was measured for all the runs of this investigation (*7*). However no new results were obtained in attempts to show an analogy between momentum and mass transfer in this work.

ACKNOWLEDGMENT

The authors wish to express their thanks to the Dow Chemical Company for the financial support of part of this work.

NOTATION

a = $\sqrt{1 + 4R_1/N_{Pe_L}}$, constant
b = constant
c = concentration of naphthalene, lb. mole/cu. ft.
c_s = saturation concentration of naphthalene
D = molecular diffusivity, sq. ft./sec.
D_p = effective pellet diameter, ft.
D_r = diameter of packed bed, ft.
E = total diffusivity, sq. ft./sec.
E_t = turbulent diffusivity, sq. ft./sec.
E'_r, E'_θ, E'_z = total diffusivities based on the area in a packed bed, sq. ft./sec.
f = $1 - P$
f_1 = value of f at $z = L$
G = superficial mass velocity, lb./hr.-sq. ft.
j_d = mass transfer factor for a packed bed = $(k_{g_{av}} RT \rho/G_{av})(\mu/\rho D)^{2/3}$
j'_d = j_d not corrected for axial mixing
j_{d_s} = j_d for a single sphere
k_c = mass transfer coefficient, ft./sec.
k_g = mass transfer coefficient, lb. mole/hr.-sq. ft.-atm.
k'_g = k_g not corrected for axial mixing
L = length of bed, ft.
N_{Pe_L} = $N_{Pe_z} L/D_p$
N_{Pe_z} = axial Peclet number, $u_s D_p/E'_z$
N_{Re} = Reynolds number, $D_p G_{av}/\mu$
N_{Sc} = Schmidt number, $\mu/\rho D$
n = number of perfect mixers
P = concentration variable, $p/p^* = c/c_s$
p = partial pressure of naphthalene
p^* = saturation partial pressure of naphthalene
R = radius variable, r/r_o; gas constant
R_1 = $6(1-\epsilon)_{av}\ RT\, k_{g_{av}}\ L/D_p u_{s_{av}}$
r = radius variable, ft.
r_o = radius of packed bed, ft.
S_v = specific surface, ft.$^{-1}$
T = temperature, °R.
t = time, sec.
u = velocity, ft./sec.
$u_s/u_{s_{av}}$ = point velocity ratio, dimensionless, Schwartz and Smith (*31*)
u_s = superficial velocity, ft./sec.
u_r, u_θ, u_z = velocity components, ft./sec.
y = mole fraction of naphthalene
z = length variable, ft.
Z = length variable, z/L

Greek Letters

θ = angle variable, radians
ϵ = void fraction
ρ = density, lb./cu. ft.
μ = viscosity, lb./ft.-sec.
σ = standard deviation from best straight line

The subscript av used with k_g, G, ϵ, u_s, etc. refers to an average value over the radius of the packed bed.

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