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LONGITUDINAL DISPERSION IN BEDS
OF FINELY DIVIDED SOLIDS

BERKELEY, CALIFORNIA

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Jerry Max Robinson

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ABSTRACT

Experimental determinations of the longitudinal dispersion coefficient and the Peclet number were made for three binary gas systems. The experimental technique involved measuring the dispersion of a step function input to a column packed with glass beads.

The diffusional model was employed to obtain the differential equation describing longitudinal dispersion in a packed column. This equation was solved with suitable boundary conditions, and the solution was used in the analysis of the experimental dispersion data.

The experimental results show a variation in the Peclet number from 0.1 to 0.6, and in the eddy dispersion coefficient from 0.03 to 1.0 cm²/sec for the Reynolds number range from 0.03 to 1.0.

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INTRODUCTION

The analysis of problems concerning fluid flow through packed columns often involves assumption that the molecules of the fluid pass through the column with no longitudinal dispersion taking place. However, because of the random motion of the molecules and macroscopic mixing from turbulence, such dispersion does occur. The effect of this is to reduce the heat- and mass-transfer driving potentials to values below those expected from the "plug flow" assumption. In the design of packed extraction columns, gas-liquid partition chromatography columns, ion-exchange columns, etc., this dispersion results in a reduction in column efficiency that cannot be calculated without further knowledge of longitudinal dispersion phenomena.

Gaseous systems at high Reynolds numbers have been investigated by McHenry and Wilhelm,¹⁴ who found that for Reynolds numbers between 10 and 400 the Peclet number is approximately constant at 1.88. This value agrees closely with the estimated upper limit of 2 for the Peclet number.^{1,4,14,15} A few gas system runs were made by Carberry and Bretton⁴ in the low-Reynolds-number range, but the data were scattered and seem inconclusive.

Liquids flowing through various packing materials have been studied by using pulse, step, and sinusoidal input functions by the following investigators:

Ebach and White⁷ used pulse inputs at low flow rates and sinusoidal inputs at high flow rates to measure Peclet numbers over a Reynolds number range of 0.01 to 200.

A pulse function input was used by Carberry and Bretton, who examined the Reynolds number range from 1 to 400. The Peclet numbers determined in their investigation show effects of bed length, particle size, and particle shape.

Strang and Geankoplis¹⁷ gathered data in the Reynolds number range from 5 to 20 by the use of sinusoidal input functions. Although the data are scattered and the Peclet number is found to depend on bed length, the results agree fairly well both with Carberry and Bretton and with Ebach and White.

The investigators mentioned above based their mathematical analysis on the diffusional model. Both E. J. Cairns⁵ and Jacques, Cotter, and Vermeulen¹⁰ employed a statistical model originally developed by H. A. Einstein.⁸ They also used step-function inputs of salt solution, and analysis based on electrical conductivity.

The object of this investigation was to relate longitudinal dispersion coefficients, for gaseous systems at low Reynolds numbers, to the system variables.

THEORY

In the development of the theory of longitudinal dispersion, there have been three mathematical models proposed. The most widely used of these is the diffusional model, which is described below. The other two are the cell-mixing model and the random-walk model.

The cell-mixing model is based on an analogy between a packed bed and a series of mixing vessels. The packed bed is considered to be composed of interconnected mixing cells. Although the mixing-cell model has been analyzed,¹ the results are difficult to use because they contain parameters that can not be readily evaluated, i.e., cell-mixing efficiency, length of a perfect mixing cell, etc.

The random-walk model is based on the analysis by H. A. Einstein of the motion of pebbles in a flowing stream. It is assumed that a molecule of a fluid flowing through a packed bed undergoes a series of "motion phases," in which it moves with the fluid, and "rest phases," in which it is stagnant. The statistical analysis of this motion yields results that can readily be used in the reduction of experimental data.^{5,10}

The diffusional model, which was used in this study, is based on the assumption that transport of a fluid through a packed column takes place by three mechanisms: convective flow, molecular diffusion, and macroscopic mixing due to turbulence. This last is assumed to occur in a manner analogous to molecular diffusion, and an equation similar to Fick's Law of Diffusion is used to define an eddy dispersion coefficient, E . E is defined by

$$N_e = -E \frac{\partial C}{\partial x}$$

where N_e = mass flux due to eddy dispersion,
 E = eddy dispersion coefficient,
 C = concentration of solute gas,
 x = distance from the column entrance.

It is also assumed that the interstitial velocity V is a constant independent of position in the bed and equals V_0/ϵ , where V_0 is superficial velocity based on the cross sectional area of the empty column and ϵ is the void fraction of the column packing. Concentration, then, is a function only of distance from the column entrance and time, e.g., $C = f(x, t)$.

Based on the above assumptions, a material balance over a short section of the column yields the differential equation

$$D \frac{\partial^2 C}{\partial x^2} - V \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \quad (1)$$

where D = longitudinal dispersion coefficient, $D_m + E$,
 t = time.

In order to solve this differential equation it is necessary to have three boundary conditions, two in space and one in time. In this analysis we are interested in the introduction of either a pulse function or a step function of a gas into a packed column in which a different gas is flowing. Obviously, then, the time boundary condition is $C(x, 0) = 0$ for all x , and one of the space boundary conditions is

determined by the charging function at $x = 0$. For a step function input of material whose solute concentration is C_0 , the charging function is $C(0, t) = \begin{matrix} 0, & t \leq 0 \\ C_0, & t > 0 \end{matrix}$. For a pulse function input of a quantity of solute q , the charging function is

$$A \epsilon \int_0^{\infty} (V C - D \frac{\partial C}{\partial x}) dt = q$$

where A is the cross-sectional area of the empty column.

The final boundary condition is not so obvious, however. If the finite column of length L may be considered equivalent to a length L of an infinite column, then

$$\lim_{x \rightarrow \infty} C(x, t) = 0$$

is a valid boundary condition. Otherwise, the boundary condition

$$\frac{\partial C(L, t)}{\partial x} = 0,$$

as derived by Danckwerts,⁶ may be used. In order for the Danckwerts boundary condition to be valid, the ratio of the longitudinal dispersion coefficients immediately outside the column and immediately inside the column must approach zero. At high fluid velocities the eddy-dispersion coefficient inside the column is much greater than that outside, and the Danckwerts assumption is reasonable. At low fluid velocities, however, the longitudinal dispersion coefficients, both inside and outside the column, approach the molecular diffusion coefficient, and their ratio approaches unity. In this case the solution resulting from the Danckwerts boundary condition is not valid. Since this investigation is concerned with low fluid velocities, the infinite column assumption is used.

The solutions of equation (1) derived from the infinite-column assumption, for step function and pulse function inputs, respectively, are

$$\frac{C}{C_0} = \frac{1}{2} \left[e^s \operatorname{erfc} \left(\frac{1+s\eta}{2\sqrt{\eta}} \right) + \operatorname{erfc} \left(\frac{1-s\eta}{2\sqrt{\eta}} \right) \right] \quad (2)$$

and

$$C = \frac{q}{AL\epsilon} \left[\frac{1}{\sqrt{\pi\eta}} e^{-\frac{(1-s\eta)^2}{4\eta}} - \frac{s}{2} e^s \operatorname{erfc} \left(\frac{1+s\eta}{2\sqrt{\eta}} \right) \right] \quad (3)$$

where $s = \frac{VL}{D}$ and $\eta = \frac{Dt}{L^2}$. For details of solutions see Appendix I.

Alternative solutions are obtained from consideration of the diffusional model with different boundary conditions,⁹ the cell-mixing model,¹ and the random-walk model.⁵ In all cases, however, the solutions converge toward a Gaussian distribution as the length of the column is increased. For short columns the solutions are appreciably different, but experimental difficulties arising due to end effects make it difficult to determine which solution fits the physical situation most precisely. To compare the solutions of equation 1, obtained by various investigators, the IBM-650 computer was programmed to calculate concentration versus time curves from each solution. This comparison is shown in Appendix VI. This investigation was confined to long columns in which there is no appreciable difference in the various solutions.

The solution of Eq. (1), then, approaches a Gaussian distribution.¹³ Therefore the peaks of the pulse function experiments, at $C = C_{\max}$, and the midpoints of the step function experiments, at $\frac{C}{C_0} = 0.5$, occur at $s\eta = 1$.

APPARATUS

The experimental equipment consisted of a gas supply, pressure-measurement and -regulation devices, an input function system (pulse or step), a packed column, a thermal conductivity (TC) cell, a recording potentiometer, and a flow-rate-measuring device. A schematic diagram of the experimental apparatus is shown in Fig. 1.

Gas was taken from pressurized gas cylinders equipped with pressure-reducing valves for rough output-pressure adjustment. The gases used were helium, air, nitrogen, and ethylene. Their specifications are listed in Appendix II.

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Fig. 1. Schematic diagram of experimental equipment.

- (a) compressed-gas cylinder
- (b) pressure control
- (c) pressure gauge
- (d) thermal conductivity cell
 - (1) reference side
 - (2) sample side
- (e) input-function device
- (f) column packed with glass beads
- (g) recording potentiometer
- (h) soap-film meter (burette).

Fine pressure regulation was accomplished with a Manostat Corporation No. 6 manostat and a National Welding Equipment Company No. 708-G pressure regulator. The column inlet and outlet pressure were measured with an Ascroft No. 1082A gauge and a U. S. Gauge No. 10763-1, respectively, each with a range of 0 to 100 psig.

The input function system for the step function was simply a 1/8-inch pipe tee with quick opening valves on two ends of the tee; the other end led to the column entrance. This system is shown in Fig. 2. The pulse function device was essentially a system of interconnected sliding chambers. It is shown in Figs. 3 and 4 to facilitate visualizing its operation. In Position 1 the gas flows directly from the reference side of the TC cell into the column. In this position the sample tube was filled with the component to be charged. The piston was then pushed to Position 2. This diverted the gas so that it flowed through the sample tube and charged the sample to the column. Construction was of copper, and the interchamber seals were rubber O rings. The sample tube was 1/4-inch copper tubing of suitable length to produce the sample volume desired.

The packed columns were 3/8-inch copper refrigerator tubing with a wall thickness of 0.032 inch. They were packed with 20-40 mesh (U.S. Standard screens) glass pavement-marking beads obtained from Minnesota Mining and Mfg. Company. Average particle diameter was 0.0246 inch. The columns were packed by inserting a small, porous glass wool plug in one end, suspending a column vertically, and slowly pouring the beads into the column. During the packing operation, the columns were vibrated by tapping to insure random packing with no trapped air pockets, and the other end was plugged with glass wool. This packing had a void fraction of 0.359. All columns had the same inside diameter (0.311 inch), packing diameter (0.0246 inch), and packing void fraction (0.359). The only parameter that varied was column length.

Analysis of the gas stream was accomplished with a Gow Mac model 9285 thermal conductivity cell. Adjustments were provided to vary the cell current from 0 to 150 milliamperes. Current was drawn from a standard 6-volt battery.

ZN-2455

Fig. 2. Step function input system.

ZN-2454

Fig. 3. Pulse-function device showing outer cylinder and charging piston.

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Fig. 4. Schematic diagram of pulse-function device.
(a) sample inlet
(b) reference gas from TC cell
(c) column entrance
(d) sample exhaust
(e) sample tube
(f) rubber O-ring seals.

The signal from the TC cell, resulting from a change in concentration (thermal conductivity), was recorded on a Leeds and Northrup Speedomax recording potentiometer. The range of the recorder was 0 to 10 millivolts, but attenuation of the TC cell signal was provided to make full-scale recorder readings represent from 10 to 10,000 millivolts.

To measure the very low gas flow rates (5 to 100 ml/min) used in this work, a burette was used as a soap-film meter. A soap-film meter is extremely accurate and gives virtually no back pressure.¹² The flow rate was controlled with an Ohio Chemical and Surgical Equipment needle valve No. 304-5208-800 having a long tapered valve stem for precise control.

PROCEDURE

The procedure followed in making experimental determinations for a step function input was as follows:

(a) The valves on the gas cylinders were opened, and the pressure adjusted with the pressure regulators and manostat to give the desired operating pressure.

(b) The snap-open valves were positioned so that only the reference gas was flowing, and the gas-flow rate through the column was adjusted with the needle valve.

(c) The TC Cell was turned on and the cell current adjusted to either 100 or 120 milliamperes. This number is rather arbitrary and affects only the sensitivity of the cell.¹²

(d) The recorder was turned on and balanced to give a zero voltage reading.

(e) The following parameters were recorded: date, gas system used, column pressure, column length, room temperature, and cell current.

(f) The positions of the quick-opening valves were reversed to produce a step function, and a start mark was made on the recorder chart. If this changed the recorder zero, balancing was repeated.

(g) The gas-flow rate was measured with the soap-film meter by introducing a drop of soap solution into the burette and timing the rise

of the resultant bubble through a given volume. The volume of gas displaced and the displacement time were recorded. This step was repeated frequently throughout the run and the average value was used in the calculations.

(h) A permanent record showing the response of the TC cell as a function of time was produced by the recording potentiometer.

For a pulse function input, Step f was changed to:

(f-1) With the charging piston in Position 1, the sample tube was filled with the gas to be used for the pulse. The piston was pushed to Position 2 to produce the pulse, and a start mark was made on the recorder chart.

CALCULATIONAL METHOD

Data reduction was accomplished by making use of the fact that for large s values the peaks of the pulse-function experiments, at $C = C_{\max}$, and the midpoint of the step-function experiments, at $\frac{C}{C_0} = 0.5$, are at $s\eta = 1$. Considering this, Eqs. (2) and (3), respectively, were simplified and solved for s to get

$$s = 4\pi \left(\frac{\frac{d C/C_0}{d t/t_{.5}}}{s\eta=1} \right)^2 = 4\pi \left(\frac{t_{.5}}{y_0} \frac{dy}{dt} \right)_{s\eta=1}^2 \quad (4)$$

and

$$s = 4\pi \left(\frac{A L \epsilon C_{\max}}{q} \right)^2 \quad (5)$$

A typical recorder-response curve is shown in Fig. 5 for a step-function input. Since the raw data are in voltage versus time, and Eq. (4) is in terms of mole fraction and time, we must make use of a calibration curve for voltage versus mole fraction, (see Appendix III).

The time at which $s\eta = 1$, t_{mid} , is easily determined, since it corresponds to $\frac{y}{y_0} = 0.5$. Knowing r_0 and r versus y , then, we can determine y_0 and obtain r at $\frac{y}{y_0} = 0.5$. When r versus y is a straight line, this is at $\frac{r}{r_0} = 0.5$.

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The time required for the breakthrough curve to flow from the column exit to the TC cell is t_f , and

$$t_{.5} = t_{mid} - t_f . \quad (6)$$

Since $\frac{dr}{dt}$ is known and $\frac{dy}{dt}$ is desired for use in Eq. (4), we make use of the relation

$$\frac{dy}{dt} = \frac{dr}{dt} \frac{dy}{dr} . \quad (7)$$

Equation (4) becomes

$$s = 4 \pi \left(\frac{t_{.5}}{y_0} \frac{dr}{dt} \frac{dy}{dr} \right)_{s\eta=1}^2 , \quad (8)$$

and $\frac{dy}{dr}$ can be obtained from y versus r . When y versus r is a straight line over the region of interest, we have

$$\frac{y}{y_0} = \frac{r}{r_0} , \quad (9)$$

and Eq. (8) reduces to

$$s = 4 \pi \left(\frac{t_{.5}}{r_0} \frac{dr}{dt} \right)_{\frac{r}{r_0} = 0.5}^2 \quad (10)$$

A typical recorder response curve is shown in Fig. 6 for a pulse function input. Equation (5) is used to calculate s .

The appropriate r -versus- y curve is used to evaluate y_{max} from r_{max} , and C_{max} is obtained from

$$C_{max} = \frac{P y_{max}}{R T} .$$

The value of q can be determined by two methods. The first method is to calculate the volume of the sample system from the physical dimensions and use

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Fig. 6. A typical recorder-response curve for a pulse-function input.

$$q = \frac{P V}{R T} .$$

The second method is based on the fact that the area under the response curve is determined by the amount of sample injected,

$$q = A V_e \int_0^{\infty} C \, dt .$$

The q value used in the calculations (7.30×10^{-7} lb mole) was obtained from the dimensions of the sample system, and was found to agree satisfactorily with an approximate value (7.6×10^{-7} lb mole) obtained by averaging the results of numerical integration of recorder-response curves. Simpson's method was used for this integration.

The void fraction of the column packing was determined by packing a graduated cylinder with the column packing and measuring the amount of water required to fill the void volume. The void fraction obtained in this manner was 0.359. The void volume could also be determined from the time required for the pulse or step function to pass through the column. The result obtained by this method fluctuated somewhat, but the average value of 0.362 compares very well to the other method.

In the determination of the gas flow rate with the soap-film meter, the presence of water vapor in the gas was neglected, since the amount present was unknown. The maximum error possible, which would occur if the gas were saturated, is less than 3%.

After determination of the s value, as described above, the longitudinal dispersion coefficient and the Peclet number can readily be calculated, since $D = \frac{VL}{s}$ and $Pe = \frac{sd}{L} \frac{p}{L}$. The eddy diffusivity is obtained from D and D_m , since

$$E = D - D_m . \quad (11)$$

When values of the molecular diffusion coefficients for the binary gases were not available in the literature, they were estimated by using the Hirschfelder, Curtiss, and Bird method with the correction proposed by Wilke and Lee.¹⁶ The estimated values for D_m at 10 psig and 297° K were

for helium-air, $D_m = 0.425 \text{ cm}^2/\text{sec}$,

for air-ethylene, $D_m = 0.100 \text{ cm}^2/\text{sec}$.

Reid and Sherwood¹⁶ gave $D_m = 0.0970 \text{ cm}^2/\text{sec}$ for nitrogen-ethylene at the same conditions.

The densities of gas mixtures were calculated by using mole fraction weighted averages. Viscosities of gas mixtures were calculated by the method of Bromley and Wilke.³

A sample calculation is included in Appendix IV.

RESULTS AND DISCUSSION

The experimentally determined curve of C versus t for run number 106, which used a step-function input, and the corresponding curve calculated from Eq. (2) are shown in Fig. 7. Since the two curves agree quite closely, the diffusional model employed is believed to be an adequate representation of the physical situation, and the equations developed from it are believed valid.

As a test of experimental technique, an equivalent series of runs was made using step- and pulse-function inputs. The results obtained by using these two different experimental techniques (pulse- and step-function input), shown in Fig. 8 as Pe versus Re , are almost the same; therefore, they are independent of the experimental technique employed. With the exception of this test, all data reported herein were taken by using a step function input.

It was found that the longitudinal diffusivity was a function of column length for columns shorter than 20 feet. Figure 9 shows this effect for the air-ethylene system. This was probably due to end effects at the column entrance and exit, which caused turbulent mixing. For long columns (20 to 40 feet) this mixing was negligible in comparison with the dispersion taking place inside the column. Only results obtained by using the 20- and 40-foot columns have been used herein for final interpretation of the data.

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Fig. 7. Comparison of experimental and calculated C-vs-t curves for run No. 106.

— experimental result
o points calculated from Eq. (2).

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Fig. 8. Comparison of pulse- and step-function inputs.
Key: $\circ$ Step-function input
 $\square$ pulse-function input
helium-air system
10-ft column.

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Fig. 9. Effect of column length on apparent Peclet number.
 Key: $\blacktriangle$ 10-foot column
 $\circ$ 20-foot column
 $\square$ 40-foot column
 air-ethylene system.

In order to minimize the possibility of mixing by natural convection, the systems used in this study were chosen so that the density difference between components was small. For the air-ethylene and nitrogen-ethylene systems the density differences (4.45×10^{-3} lb/ft³ and 0 lb/ft³, respectively) were very small and the pure gases were used. For the helium-air system, however, the density difference between the pure components was 0.1075 lb/ft³. Consequently, a mixture of air in helium was made for which $y_{\text{air}} = 0.146$, and this mixture was used as one component of the system. The resulting density difference was 0.0157 lb/ft³.

In the past, longitudinal dispersion data generally have been presented by plotting dimensionless groups characteristic of the dispersion process and the turbulence induced by the flow of the fluid. The group chosen to characterize turbulence has been the Reynolds number,

$$Re = \frac{d_p \rho V_o}{\mu} \quad (12)$$

and the group chosen to characterize dispersion has been the Peclet number,

$$Pe = \frac{d_p V}{D} \quad (13)$$

The results of this work are shown in this manner in Fig. 10. Figure 11 shows the results of this work together with those of other recent investigators.

The gas data of Carberry and Bretton⁴ were obtained by injecting a pulse of air into a helium stream. The column used was a "gas chromatographic column several meters in length." The properties of the packing used in the column were not presented. To get their data in terms of the coordinates of Fig. 11, it was necessary to assume a void fraction for their column packing. The packing usually used in chromatographic columns is crushed firebrick, and a representative value of the porosity of this packing is 0.6.⁹ Incidentally, if the data from this investigation are plotted in the same manner as those presented by Carberry and Bretton,

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Fig. 10. Peclet number as a function of Reynolds number.

Key: $\circ$ helium-air system
 $\bullet$ air-ethylene system
 $\square$ nitrogen-ethylene system.

Fig. 11. Comparison with other data as Peclet number versus Reynolds number.

(a) This work	(gas)
(b) Carberry and Bretton	(gas)
(c) McHenry and Wilhelm	(gas)
(d) Ebach and White	(liquid)
(e) Strang and Geankoplis	(liquid)
(f) Carberry and Bretton	(liquid).

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the relative positions of the two curves are the same. The lower Peclet numbers obtained by Carberry and Bretton might be expected owing to the dispersive effect of diffusion within the porous packing,¹⁷ and possibly free convection effects due to the large density difference when the air pulse was injected into the helium stream.

Intuitively, it would seem that the Peclet number should increase with increasing Reynolds number; however, the Peclet numbers in Fig. 10 reach a maximum and then decrease. This is believed to be due to the experimental difficulty in producing a perfect pulse- or step-function input. In order for the input function to be a good approximation of a step function, its slope must be large in comparison with that of the concentration profile at the column exit. For a step function input, then, it is necessary that

$$(dc/dt)_{t=0} >> (dc/dt)_{t=t_{.5}}$$

The decrease of Pe with increase of Re occurred when this condition was not fulfilled and the input function ceased to be a good approximation of a step or pulse function. In the absence of the above limitation it is believed that the data of this investigation would extrapolate into those of McHenry and Wilhelm¹⁴ with satisfactory agreement. The data of McHenry and Wilhelm and of Ebach and White⁷ (in the high Re range) were obtained by using a sinusoidal input function that is not subject to the above limitation. Their method, however, leads to "analytical difficulty" at low Reynolds numbers.⁷

Ebach and White have studied the effect of viscosity on longitudinal mixing for liquid systems and found no variation with viscosity over the range of 1 to 26 centipoises. The Reynolds number, then, may not be an apt choice of variables to characterize the variation of the Peclet number. The cell-mixing model suggests a proportionality between the eddy dispersion coefficient and the product of particle diameter and interstitial velocity. In an attempt to generalize, Cairns⁵ has proposed that the particle diameter be replaced by a hydraulic radius, r_m , where

$$r_m = \frac{\text{void volume}}{\text{surface area}}$$

For spherical particles in a cylindrical tube we have

$$4m = \frac{D_c \epsilon}{3/2 D_c / dp (1 - \epsilon) + 1} \quad (14)$$

The data would then be represented by a plot of $4mV$ versus the eddy dispersion coefficient or a modified Peclet number, $\frac{4mV}{E}$. Representative data are shown on this type of plot in Figs. 12 and 13. The gas data of Carberry and Bretton are not shown, since the parameters necessary to convert to these coordinates are not available.

Jacques, Cotter, and Vermeulen¹⁰ have proposed alternative definitions of the Peclet and Reynolds numbers in an effort to account for particle size and shape effects which they found in their investigation. These definitions are

$$Pe' = Pe \epsilon = \frac{d_p V_o}{E} \quad (15)$$

and

$$Re' = \frac{Re}{1 - \epsilon} = \frac{d_p \rho V_o}{\mu (1 - \epsilon)} \quad (16)$$

This type of correlation is shown in Fig. 14.

Experimental data and results are presented in tabular form in Appendix V.

SUMMARY

Experimental determinations of the Peclet number and longitudinal dispersion coefficient were made for three gas systems. The analysis of the experimental data was based on the solution of the differential equation resulting from the use of the diffusion model.

The experimental technique employed a thermal conductivity cell to measure the degree of dispersion of a step function due to passage through a column packed with glass beads.

The results of this investigation show a variation in the Peclet number from 0.1 to 0.6, and in the eddy dispersion coefficient from 0.03 to 1.0 cm²/sec for the Reynolds number range from 0.03 to 1.0.

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Fig. 12. Comparison with other data given as E vs $4mV$ (after Cairns).

(a) This work	(gas)
(b) McHenry and Wilhelm	(gas)
(c) Strang and Geankoplis	(liquid)
(d) Carberry and Bretton,
and Ebach and White	(liquid)
(e) Cairns	(liquid).

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Fig. 13. Comparison with other data given as $4m V/E$ vs $4m V$ (after Cairns).

(a) This work	(gas)
(b) McHenry and Wilhelm	(gas)
(c) Strang and Geankoplis	(liquid)
(d) Carberry and Bretton,
and Ebach and White	(liquid)
(e) Cairns	(liquid).

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Fig. 14. Comparison with the data of Jacques, Cotter, and Vermeulen.

Key: $\circ$ helium-air system
 $\bullet$ air-ethylene system
 $\square$ nitrogen-ethylene system
 $\triangle$ and — Jacques, Cotter, and Vermeulen.

APPENDIX I

Solution of Differential Equation

The differential equation to be solved is

$$D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t}, \quad (A 1)$$

with two of the boundary conditions being

$$\begin{aligned} C(x, 0) &= 0 \text{ for all } x, \\ \lim_{x \rightarrow \infty} C(x, t) &= 0 \text{ for all } t. \end{aligned}$$

For a Step-Function Input

The third boundary condition is

$$C(0, t) = \begin{cases} 0, & t \leq 0 \\ C_0, & t > 0 \end{cases}$$

Upon assumption of a solution of the form

$$C = \psi \exp \left(\frac{v x}{2 D} - \frac{v^2 t}{4 D} \right),$$

Eq. (A 1) can be reduced to

$$D \frac{\partial^2 \psi}{\partial x^2} = \frac{\partial \psi}{\partial t}, \quad (A 2)$$

with boundary conditions

$$\begin{aligned} \psi(x, 0) &= 0 \text{ for all } x \\ \lim_{x \rightarrow \infty} \psi(x, t) &= 0 \text{ for all } t, \\ \psi(0, t) &= C_0 \exp \left(\frac{v^2 t}{4 D} \right). \end{aligned}$$

This system of equations can be solved by using the Laplace transformation,

$$\bar{\psi}(x) = \int_0^{\infty} e^{-pt} \psi(x, t) dt.$$

Equation (A 2) becomes

$$D \frac{d^2 \bar{\psi}}{dx^2} = p \bar{\psi} - \psi(x, 0+) = p \bar{\psi} \quad (A 3)$$

with boundary conditions

$$\bar{\psi}(\infty) = 0, \quad (A 4)$$

$$\bar{\psi} = C_0 \left(\frac{1}{p - v^2/4D} \right). \quad (A 5)$$

The solution of (A 3) is

$$\bar{\psi} = A \exp(-\sqrt{p/D} x) + B \exp(\sqrt{p/D} x). \quad (A 6)$$

Applying Eq. (A 4), we find

$$B = 0, \\ \bar{\psi} = A \exp(-\sqrt{p/D} x).$$

Applying Eq. (A 5), we find

$$A = C_0 \left(\frac{1}{p - v^2/4D} \right).$$

Therefore

$$\bar{\psi} = C_0 \left(\frac{1}{p - v^2/4D} \right) \exp(-\sqrt{p/D} x). \quad (A 7)$$

The inverse transform of Eq. (A 7) is²

$$\psi = \frac{C_0}{2} \left[\exp\left(\frac{V^2 t + 2 V x}{4 D}\right) \operatorname{erfc}\left(\frac{x}{2 \sqrt{D t}} + \frac{V}{2} \sqrt{t/D}\right) + \exp\left(\frac{V^2 t - 2 V x}{4 D}\right) \operatorname{erfc}\left(\frac{x}{2 \sqrt{D t}} - \frac{V}{2} \sqrt{t/D}\right) \right],$$

which gives

$$\begin{aligned} \frac{C}{C_0} = \frac{1}{2} & \left[\exp\left(\frac{V^2 t + 2 V x}{4 D}\right) \operatorname{erfc}\left(\frac{x + V t}{2 \sqrt{D t}}\right) + \exp\left(\frac{V^2 t - 2 V x}{4 D}\right) \operatorname{erfc}\left(\frac{x - V t}{2 \sqrt{D t}}\right) \right] \\ & \exp\left[-\left(\frac{V^2 t - V x}{4 D}\right)\right]. \end{aligned}$$

Let $x = L$, $s = VL/D$, and $\eta = Dt/L^2$; then

$$\frac{C}{C_0} = \frac{1}{2} \left[e^s \operatorname{erfc}\left(\frac{1 + s\eta}{2 \sqrt{\eta}}\right) + \operatorname{erfc}\left(\frac{1 - s\eta}{2 \sqrt{\eta}}\right) \right],$$

where $\operatorname{erfc} Z = 1 - \operatorname{erf} Z = 1 - \frac{2}{\sqrt{\pi}} \int_0^Z e^{-u^2} du$.

For a Pulse-Function Input

The third boundary condition is

$$A \in \int_0^\infty (V C - D \frac{\partial C}{\partial x}) dt = q.$$

The solution follows the same procedure except that Eqs. (A 3), (A 4), and (A 5) become

$$D \frac{d^2 \bar{\psi}}{dx^2} = p \bar{\psi},$$

$$\bar{\psi}(\infty) = 0, \quad (A 8)$$

$$\frac{V}{2} \bar{\psi} - D \frac{d \bar{\psi}}{dx} = \frac{q}{A \epsilon}. \quad (A 9)$$

Equation (A 6) is the same, and using (A 8) and (A 9), we find,

$$\bar{\psi} = \frac{q}{A \epsilon} \left(\frac{1}{V/2 + \sqrt{p/D}} \right) \exp(-\sqrt{p/D} x). \quad (A 10)$$

The inverse transform of (A 10) is²

$$\psi = \frac{q}{A \epsilon} \left[\frac{1}{\sqrt{\pi Dt}} \exp\left(-\frac{x^2}{4Dt}\right) - V/2D \exp\left(\frac{V^2 t + 2Vx}{4D}\right) \operatorname{erfc}\left(\frac{x + Vt}{2\sqrt{Dt}}\right) \right],$$

which gives

$$C = \frac{q}{A \epsilon} \left[\frac{1}{\sqrt{\pi Dt}} \exp\left(-\frac{x^2}{4Dt}\right) - V/2D \exp\left(\frac{V^2 t + 2Vx}{4D}\right) \operatorname{erfc}\left(\frac{x + Vt}{2\sqrt{Dt}}\right) \right] \exp\left(\frac{2Vx - V^2 t}{4D}\right),$$

or

$$C = \frac{q}{A L \epsilon} \left[\frac{1}{\sqrt{\pi \eta}} \exp\left[-\frac{(1 - s\eta)^2}{4\eta}\right] - s/2 e^s \operatorname{erfc}\left(\frac{1 + s\eta}{2\sqrt{\eta}}\right) \right].$$

APPENDIX II

Properties of Gases

Helium

U. S. Navy, grade AA

purity	99.99%
molecular weight	4.00
density at 297° K, 24.7 psia	0.01721 lb/ft ³
viscosity at 297° K	0.0448 lb/ft-hr

Ethylene

Matheson, C.P. grade

purity	99.5%
molecular weight	28.0
density at 297° K, 24.7 psia	0.1224 lb/ft ³
viscosity at 297° K, 24.7 psia	0.0244 lb/ft-hr

Nitrogen

National Cylinder and Gas, prepurified

purity	99.99%
molecular weight	28.0
density at 297° K, 24.7 psia	0.1224 lb/ft ³
viscosity at 297° K	0.0426 lb/ft-hr

Air

Liquid Carbonic

molecular weight	29.0
density at 297° K, 24.7 psia	0.1247 lb/ft ³
viscosity at 297° K	0.0433 lb/ft-hr

APPENDIX III

Calibration of Thermal-Conductivity Cell

In order to convert the response of the thermal conductivity cell from voltage to concentration, calibration runs were made under conditions similar to the operating conditions of the column. The voltage output of the TC cell, resulting from comparison of a mixture of known composition with the reference gas, was measured. The calibration curves are shown in Figs. 15, 16, and 17.

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Fig. 15. Mole fraction air in helium-air mixture versus response of thermal-conductivity cell.

MU-20185

Fig. 16. Mole fraction ethylene in ethylene-air mixture versus response of thermal conductivity cell.

MU-20186

Fig. 17. Mole fraction ethylene in ethylene-nitrogen mixture versus response of thermal conductivity cell.

APPENDIX IV

Sample Calculation for Run No. 106

Constants

Column area, A	$5.28 \times 10^{-4} \text{ ft}^2$
Column pressure, P	24.7 psia
Length, L_c , of connector from column exit to TC cell	0.60 ft
Connector area, A_c	$1.97 \times 10^{-4} \text{ ft}^2$
Void fraction of packing, ϵ	0.359
Particle diameter, d_p	$2.05 \times 10^{-3} \text{ ft}$
Gas density, ρ	$1.014 \times 10^{-3} \text{ lb/ft}^3$
Gas viscosity, μ	$4.64 \times 10^{-2} \text{ lb/ft-hr}$
Column length, L	20 ft

Experimental Data

Gas-flow rate at 1 atm, v	26.2 ml/min
Time to $r/r_o = 0.5$, t_{mid}	7.14 min
Slope at $r/r_o = 0.5$, $(dr/dt)_{0.5}$	31.5 mv/min
Maximum recorder response, r_o	19.35 mv

Calculations

Correction for flow time through column-TC cell connector:

$$t_f = L_c/v (A_c) (P/14.7) (\text{unit conversion factors}),$$

$$t_f = 0.60/26.2 (1.97 \times 10^{-4}) (24.7/14.7) (1000) (28.3) = 0.21 \text{ min},$$

$$t_{.5} = t_{mid} - t_f = 7.14 - 0.21 = 6.93 \text{ min}.$$

Gas-flow velocity:

$$V_o = v/A (14.7/P) (\text{unit conversion factors})$$

* Gas properties for use in calculation of the Reynolds number were evaluated at $C/C_o = 0.5$.

$$V_o = 26.2/5.28 \times 10^{-4} (14.7/24.7) \left[\frac{60}{1000 (28.3)} \right] = 62.7 \text{ ft/hr} ,$$

$$V = V_o/\epsilon = 62.7/0.359 = 174.6 \text{ ft/hr}.$$

Reynolds number:

$$Re = d_p \rho V_o / \mu = \frac{2.05 \times 10^{-3} (1.014 \times 10^{-3}) (24.7) (62.7)}{4.64 \times 10^{-2}} = 0.0695$$

4 m V:

$$4 \text{ m } V = \frac{D_c \epsilon V}{3/2 D_c / d_p (1 - \epsilon) + 1} = \frac{0.311 (0.359) (174.6)}{3/2 (0.311/0.0246) (1 - 0.359) + 1}$$

$$4 \text{ m } V = 0.1237 \text{ ft}^2/\text{hr},$$

$$4 \text{ m } V = 0.0320 \text{ cm}^2/\text{sec}.$$

s:

$$s = 4 \pi (t_{.5}/y_o \quad dy/dt)^2_{t = t_{.5}}$$

Since we have $y_o \approx 0.14$, Fig. 14 shows that the y-versus-r calibration curve is approximately a straight line over the region of interest. Therefore,

$$y/y_o = r/r_o,$$

$$1/y_o \quad dy/dt = 1/r_o \quad dr/dt ,$$

and

$$s = 4 \pi (t_{.5}/r_o \quad dr/dt)^2_{t = t_{.5}} ,$$

$$s = 4 \pi \left[6.93/19.35 (31.5) \right]^2 = 1601 .$$

Peclet number:

$$Pe = s d_p / L = 1601 (2.0 \times 10^{-3} / 20) = 0.1641.$$

Eddy-dispersion coefficient

$$E = V L/s - D_m = 174.6 (20) (.258) / 1601 = 0.425,$$

$$E = 0.137 \text{ cm}^2/\text{sec}.$$

$$\frac{4 \text{ m V}}{E} :$$

$$\frac{4 \text{ m V}}{E} = \frac{0.0320}{0.137} = 0.234 .$$

APPENDIX V

Table I. Experimental Data

$T = 297^{\circ}\text{K}$

$p = 24.7 \text{ psia}$

pressure drop $\approx 0.0014 \text{ v}$ $\frac{\text{psia}}{\text{ft. of column}}$

Run No.	Column length (ft)	Gas-flow rate at 14.7 psia (ml/min)	Time to $C/C_0 = 0.5$ (min)	$\left[\frac{dr}{dt}\right]_{C/C_0 = 0.5}$ (mv/min)	Maximum recorder response (mv)
Helium-air system ($y_0 = 0.146$)					
101	20	101	1.86	200	23.5
102	20	68.0	2.81	125	22.8
103	20	34.00	5.60	51.0	22.0
104	20	21.3	8.84	26.4	21.0
105	20	47.4	4.04	71.5	20.0
106	20	26.2	7.14	31.6	15.5
107	20	10.4	16.94	9.88	18.8
Air-ethylene system ($y_0 = 1.0$)					
151	20	96.5	1.94	253	
152	20	49.8	3.90	123	
153	20	45.8	4.25	134	
154	20	55.8	3.46	157	
155	20	55.1	2.38	195	
156	20	91.0	2.06	220	
157	20	88.0	2.24	212	
158	20	43.6	4.40	118	
159	20	26.3	7.30	68.5	
160	20	32.00	5.94	85.5	
161	20	18.1	10.33	45.6	
162	20	38.3	5.02	104	
163	20	109	1.76	253	
164	20	59.1	3.27	153	

Table I (continued)

Run No.	Column length (ft)	Gas-flow rate at 14.7 psia (ml/min)	Time to $C/C_0 = 0.5$ (min)	$\left[\frac{dr}{dt}\right]_{C/C_0 = 0.5}$ (mv/ml·min)	Maximum recorder response (mv)
165	20	32.8	5.78	89.0	
166	20	20.1	9.29	52.2	
167	20	11.4	15.77	26.2	
168	20	75.0	2.57	166	
169	20	7500	2.57	176	
201	40	98.0	3.63	170	
202	40	106	3.40	182	
203	40	72.5	5.06	123	
204	40	32.0	11.76	60.0	
Nitrogen-ethylene system ($y_0 = 1.0$)					
251	40	98.3	3.48	186	27.2
252	40	83.4	4.28	170	26.5
253	40	72.9	4.95	143	25.8
254	40	58.8	6.15	108	25.5
255	40	47.0	7.79	90.0	25.2
256	40	121	2.93	233	26.2
257	40	104	3.48	186	25.5
258	40	86.3	4.24	152	25.0
259	40	59.1	6.28	114	24.5
260	40	37.8	9.99	75.0	24.5

Table II. Calculated results

Run No.	t _{.5} (min)	s	Pe	Re	E (cm ² /sec)	4mV (cm ² /sec)	4mV/E
101	1.80	2820	0.289	0.268	0.805	0.1232	0.153
102	2.73	2830	0.290	0.1803	0.400	0.0830	0.207
103	5.44	2000	0.205	0.0901	0.159	0.0415	0.261
104	8.58	1459	0.1496	0.0565	0.076	0.0260	0.342
105	3.92	2460	0.252	0.1257	0.237	0.0578	0.244
106	6.93	1601	0.1641	0.0695	0.137	0.0320	0.234
107	16.40	938	0.0961	0.0276	---	0.0127	---
151	1.88	4450	0.456	1.803	0.645	0.1178	0.182
152	3.79	4250	0.437	0.930	0.301	0.0608	0.202
153	4.13	6040	0.619	0.855	0.161	0.0559	0.347
154	3.36	5440	0.558	1.041	0.252	0.0680	0.270
155	2.28	3880	0.398	1.027	0.387	0.0672	0.174
156	2.00	3800	0.390	1.697	0.721	0.1110	0.154
157	2.18	4200	0.431	1.643	0.619	0.1073	0.174
158	4.27	4950	0.507	0.815	0.202	0.0532	0.264
159	7.09	4530	0.475	0.497	0.095	0.0321	0.339
160	5.76	4750	0.488	0.598	0.131	0.0391	0.298
161	10.02	4100	0.421	0.338	0.051	0.0221	0.432
162	4.87	5080	0.521	0.715	0.159	0.0468	0.294
163	1.71	3680	0.378	2.04	0.915	0.1330	0.145
164	3.17	4630	0.475	1.105	0.338	0.0722	0.214
165	5.61	4900	0.501	0.612	0.130	0.0400	0.308
166	9.01	4350	0.446	0.394	0.0665	0.0258	0.388
167	15.28	3140	0.322	0.213	0.0246	0.0139	0.565
168	2.50	3380	0.347	1.400	0.660	0.0915	0.139
169	2.50	3800	0.389	1.400	0.579	0.0915	0.158
201	3.57	7230	0.370	1.830	0.832	0.1195	0.144
202	3.35	7340	0.376	1.974	0.890	0.1290	0.145
203	4.98	7350	0.376	1.353	0.579	0.0885	0.153

Table II (continued)

Run No.	t _{.5} (min)	s	Pe	Re	E (cm ² /sec)	$\frac{4mV}{E}$ (cm ² /sec)	$\frac{4mV}{E}$
204	11.58	9460	0.485	0.598	0.132	0.0391	0.296
251	3.42	6840	0.350	1.813	0.891	0.1199	0.134
252	4.21	9150	0.469	1.540	0.530	0.1017	0.192
253	4.87	9230	0.473	1.342	0.445	0.0889	0.200
254	6.05	8160	0.418	1.084	0.398	0.0717	0.180
255	7.67	9390	0.481	0.869	0.247	0.0573	0.232
256	2.00	8230	0.422	2.23	0.913	0.1477	0.162
257	3.43	7850	0.402	1.920	0.813	0.1269	0.156
258	4.18	8070	0.413	1.589	0.638	0.1052	0.165
259	6.19	10460	0.535	1.091	0.292	0.0722	0.247
260	9.84	11380	0.583	0.698	0.131	0.0462	0.353

APPENDIX VI

Numerical Comparison of Solutions of Differential Equation

Equation (1) has been solved by several investigators for a pulse-function input. Some of the solutions, in the nomenclature of this work, are as follows.

This work:

$$C = \frac{q}{A L \epsilon} \left[\frac{1}{\sqrt{\pi \eta}} \exp \left[-\frac{(1 - s\eta)^2}{4 \eta} \right] - \frac{s}{2} e^s \operatorname{erfc} \left(\frac{1 + s\eta}{2 \sqrt{\eta}} \right) \right] \quad (A 11)$$

Carberry and Bretton:⁴

$$C = \frac{q}{2 A L \epsilon \sqrt{\pi \eta}} \exp \left[-\frac{(1 - s\eta)^2}{4 \eta} \right] \quad (A 12)$$

Ebach and White:⁷

$$C = \frac{q}{2 A L \epsilon s \eta \sqrt{\pi \eta}} \exp \left[-\frac{(1 - s\eta)^2}{4 \eta} \right] \quad (A 13)$$

R. H. Houston:⁹

$$C = \frac{q}{4 A L \epsilon} \left[\frac{4}{\sqrt{\pi \eta}} \left(\frac{s^2 \eta}{4} + \frac{1}{2} \right) \exp \left[-\frac{(1 - s\eta)^2}{4 \eta} \right] - 2 \left(\frac{s^3 \eta}{4} + s + \frac{s^2}{4} \right) e^s \operatorname{erfc} \left(\frac{1 + s\eta}{2 \sqrt{\eta}} \right) \right] \quad (A 14)$$

Equations (A 11) through (A 14) were numerically evaluated by using the IBM 650 computer and the approximation¹¹

$$\operatorname{erfc} x = \frac{e^{-x^2}}{x \sqrt{\pi}} \left(1 - \frac{1}{2 x^2} + \frac{1 \cdot 3}{4 x^4} - \frac{1 \cdot 3 \cdot 5}{8 x^6} + \dots \right)$$

Figures 18, 19, and 20 show concentration as a function of the dimensionless time variable, $s\eta$, with s as a parameter. The constants used in the evaluation of Eqs. (A 11) through (A 14) are as follows: $q = 7.60 \times 10^{-7}$ lb mole, $A = 5.15 \times 10^{-4}$ ft², $L = 6.90$ ft, $\epsilon = 0.410$. It is apparent from these graphs that as s increases, the difference between the various solutions decreases, and the curves approach a Gaussian distribution with $C_{\max}$ at $s\eta = 1$.

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Fig. 18. Concentration at $x = L$ versus dimensionless time for $s = 100$.

Key: — This work
- - - Carberry and Bretton
... Houston, and Ebach and White.

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Fig. 19. Concentration at $x = L$ versus dimensionless time for $s = 500$.

Key: — This work
- - - Carberry and Bretton
... Houston, and Ebach and White.

MU-20189

Fig. 20. Concentration at $x = L$ versus dimensionless time for $s = 1000$.

Key: — This work; Carberry and Bretton;
Houston; and Ebach and White.

NOMENCLATURE

A	cross-sectional area of empty column
A_c	column-TC cell connector area
C	concentration of solute gas
C_o	concentration of input step-function gas
D	longitudinal dispersion coefficient
D_c	inside diameter of column
D_m	molecular diffusion coefficient
d_p	particle diameter
E	eddy-dispersion coefficient
L	column length
L_c	column-TC cell connector length
N_e	mass flux due to eddy dispersion
P	column pressure
Pe	Peclet number, $d_p V/D$
Pe'	modified Peclet number, $d_p V_o/D$
q	quantity of gas charged in a pulse input
R	gas constant
Re	Reynolds number, $d_p \rho V/\mu$
Re'	modified Reynolds number, $Re/(1-\epsilon)$
r	recorder response
r_o	recorder response due to input step-function gas
s	dimensionless mixing parameter, VL/D
T	temperature
t	time
t_f	flow time through column-TC cell connector
t_{mid}	time at which $C/C_o = 0.5$ (uncorrected for t_f)
$t_{.5}$	time at which $C/C_o = 0.5$, $t_{mid} - t_f$

v	volumetric flow rate of gas at 1 atmosphere, ml/min
V_o	superficial velocity
V	interstitial velocity, V_o/ϵ
x	axial distance from column entrance
y	mole fraction of solute gas
y_o	mole fraction of input step-function gas
η	dimensionless parameter, Dt/L^2
ρ	density
μ	viscosity

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