Effect of Fluid Properties on Dispersion in Flow through Packed Beds

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An experimental study is described on the effect of liquid properties on axial dispersion in packed beds. Working with water between 278 and 373 K, the range $57 < Sc < 1{,}930$ was covered, and this represents a significant fraction of the domain located between what is traditionally considered "gas behavior" (that is, $Sc \cong 1$) and "liquid behavior" (typically, $Sc > 600$). A representation of Pe_L vs. Pe_m reveals the existence of a very flat minimum (typically $10^2 < Pe_m < 10^4$) for each value of Sc. The value of the minimum is almost insensitive to variations in Sc, for $Sc > 700$, but it increases significantly with a decrease in Sc, for $Sc < 700$. The experimental data used to test available correlations for predicting D_L show that they are inaccurate over considerable ranges of the variables.

Introduction

The problem of dispersion in flow through packed beds is of central importance to chemical engineers, and it is treated in some detail in textbooks on chemical reaction engineering, at least since the pioneering text of Levenspiel (1962). Hydrologists and geophysicists have had a natural interest in the topic for nearly a hundred years now (Slichter, 1905), with good coverage on the subject offered in books devoted to the study of flow through porous media [see, for example, Bear (1972) and Scheidegger (1974)]. Petroleum engineers are also concerned with the same topic under the heading of miscible displacement (Pfannkuch, 1963). This variety of fields of application has propelled the search for experimental data, with a marked peak of activity in the 1950s and 1960s. The vast majority of data on dispersion are either for air or water near ambient temperature, and some data also have been published for the flow of viscous liquids (Blackwell, 1962).

At a "macroscopic" level, the quantitative treatment of dispersion is currently based on the use of an equation analogous to Fick's law, with a dispersion coefficient used instead of a molecular diffusion coefficient; the longitudinal (or axial) dispersion coefficient D_L measures dispersion in the direction of flow, whereas dispersion perpendicular to the direction of flow is related to the transverse dispersion coefficient D_T. In this article consideration will be given only to the effect of fluid properties on D_L. [The effect of fluid properties on D_T is reported elsewhere by Delgado and Guedes de Carvalho (2001).]

In the limit of very low fluid velocity, dispersion is determined solely by molecular diffusion, with $D_L = D_m/\tau$ (τ being the tortuosity factor for diffusion). At high fluid velocities, dispersion is purely "fluid mechanical" (Wilhelm 1962), with $D_L = ud/Pe_L(\infty)$, where u is the interstitial fluid velocity and $Pe_L(\infty) \cong 2$ for gas or liquid flow through beds of (approximately) isometric particles with diameter d. A common approximation for the intermediate range of fluid velocities is to assume that the effects of molecular diffusion and fluid-mechanical dispersion are additive and the resulting expression is

$$D_L = D'_m + ud/Pe_L(\infty), \qquad (1)$$

where $D'_m = D_m/\tau$. This equation is expected to give the correct asymptotic behavior at high and low values of $Pe_m (= ud/D_m)$. For gases, this is confirmed in Figure 1, but for liquids (Figure 2) the data do not cover the extreme conditions.

However, these figures show that Eq. 1 is inaccurate over part of the intermediate range of Pe_m. In the case of gas flow, shown in Figure 1, significant deviations are observed only in the range $0.6 < Pe_m < 60$; the experimental values of $Pe_L (= ud/D_L)$ are generally higher than predicted by Eq. 1. Several equations have been proposed to represent the data in this intermediate range, and the equations of Hiby (1962)

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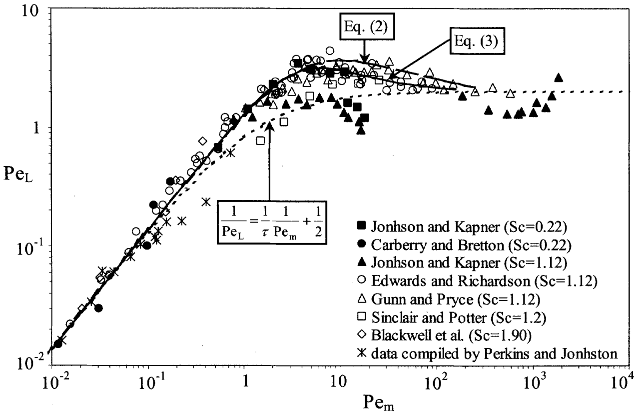

Figure 1. Longitudinal dispersion in gases.

$$\frac{1}{Pe_L} = \frac{1}{\tau Pe_m} + \frac{0.65}{1 + 7\sqrt{\dfrac{1}{Pe_m}}} \qquad (\mathrm{Re} < 100) \qquad (2)$$

and of Edwards and Richardson (1968)

$$\frac{1}{Pe_L} = \frac{0.73}{Pe_m} + \frac{0.5}{1 + \dfrac{9.7}{Pe_m}}$$

$$(0.08 < \mathrm{Re} < 50;\ 0.4 < d(\mathrm{mm}) < 6.0) \qquad (3)$$

are shown to fit the data points reasonably well.

With liquids, deviations from Eq. 1 occur over the much wider range $2 < Pe_m < 10^6$, the experimental values of Pe_L being significantly lower than predicted by that equation. It can be noted that scatter is more pronounced in the case of liquids, and it would be even more striking if we did not omit the data by Harleman and Rumer (1963), Harleman et al. (1963), Rumer (1962), and Hiby (1962), that were obtained with columns with a ratio L/D, between length and diameter, below about 10.

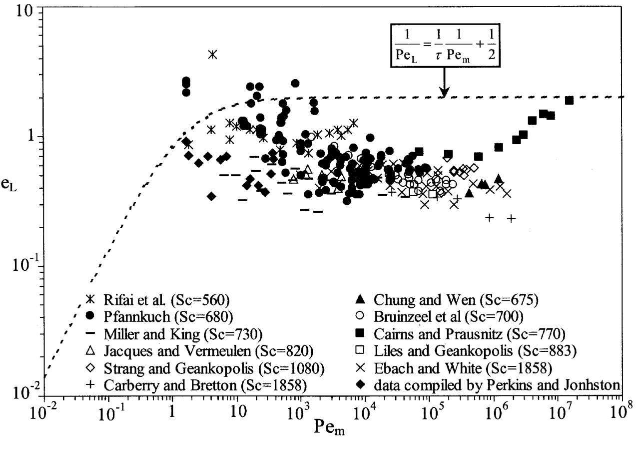

Figure 2. Longitudinal dispersion in liquids.

The difference in behavior between gases and liquids has to be ascribed to the dependence of Pe_L on $Sc(= \mu/\rho D_m)$. Indeed, dimensional analysis shows that $Pe_L = f(Pe_m, Sc)$, for beds of monosized particles of constant voidage, which is approximately the case for the majority of experiments reported in the literature.

For most gaseous mixtures, far from the critical point, the value of Sc is close to unity, whereas for most solutes in cold water, it is $550 < Sc < 2{,}000$. Curiously, no data on longitudinal dispersion seem to have been reported for the range $20 < Sc < 550$.

The present study was undertaken to obtain data in this intermediate range of Sc, by working with dilute solutions of sodium chloride in water, in the temperature range 293 K $< T <$ 373 K.

Experimental Technique

Typically, dispersion along the direction of flow is studied by following the distortion of some concentration wave (of a tracer), as it progresses along the packing. In our experiments, a step in tracer concentration was introduced at the top of a long packed bed, of constant cross section, and the variation of tracer concentration, in the stream leaving the bed, was recorded continuously.

The partial differential equation describing tracer transport in the bed is

$$D_L \frac{\partial^2 C}{\partial z^2} - u \frac{\partial C}{\partial z} = \frac{\partial C}{\partial t} \qquad (4)$$

where z measures length along the bed. For a step input (from C_0 to C_S), the concentration at the outlet of the bed ($z = L$) is known (Danckwerts, 1953) to be given by

$$\frac{C - C_0}{C_S - C_0} = \frac{1}{2}\left[1 - \mathrm{erf}\left(\frac{L - ut}{2\sqrt{D_L t}}\right)\right] \qquad (5)$$

if L is sufficiently large. This result can be written as

$$F = \frac{1}{2}\left[1 - \mathrm{erf}\left(\sqrt{\frac{LPe_L}{\theta d}}\,\frac{(1-\theta)}{2}\right)\right] \qquad (6)$$

where $F = (C - C_0)/(C_S - C_0)$ is the dimensionless concentration rise, and $\theta = t/\bar{t}$ is the dimensionless time ($\bar{t}$ being the mean residence time of fluid in the bed).

For each set of C vs. t values obtained in one experiment, the values of F vs. θ were calculated and the value of Pe_L determined to give the least deviation between the experimental points and the line representing Eq. 6. Figure 3 helps illustrate the method, and it also gives some idea about its sensitivity to variations in Pe_L.

The rig used in the majority of our experiments (including all those at temperatures above ambient) is sketched in Figure 4, but the various items are not to scale. Two large stainless-steel reservoirs (about 0.3 m in diameter and 1.0 m tall) were used to keep the distilled water and the dilute solution of sodium chloride (up to 1.5 kg/m^3 in salt) immersed in a silicone oil thermostatic bath. The use of very dilute salt solu-

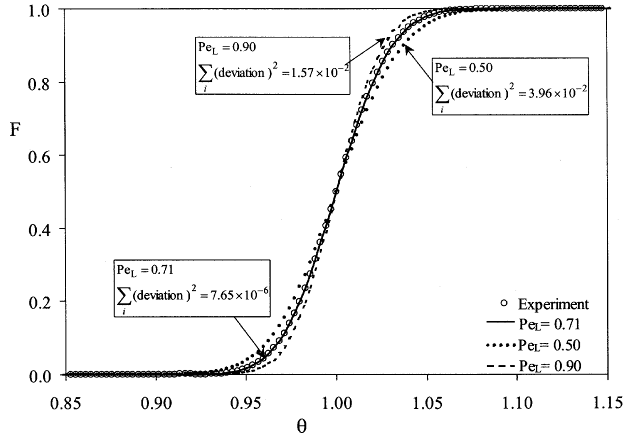

Figure 3. Comparison of experiment with Eq. 6 for three values of Pe_L.

tions and the care taken in equalizing the termperature of both liquids is needed to avoid dispersion by natural convection. Both the distilled water and the salt solution were degassed *in situ*, by bubbling under a vacuum to avoid liberation of small air bubbles inside the test column at the higher temperatures. Both reservoirs were connected (at the top) to the compressed-air line (about 4 bar) to have the rig permanently pressurized and help discharge the liquid through the flow regulating valve V, open to the atmosphere.

At the top of the test column (3.0 m long and 0.047 m in diameter, made of brass), a three-way valve (T) could be manipulated to select the feed. At the beginning of each experiment, it was turned to let distilled water flow through the bed, until no salt was detected in the conductivity meter monitoring the exit stream. After that, it was turned to allow the salt solution to go through the column, at a constant flow rate, measured by an orifice meter connected to a differential pressure transducer. The dead space between the three-way valve and the top of the bed was less than 0.1% of the void space in the bed proper, and this is important to ensure that a sharp step input of tracer is obtained in the bed. The conductivity cell at the exit of the test column was connected to a microcomputer, and values of the salt concentration were recorded at a frequency of 10 Hz. Care was taken to have a negligible volume of liquid between the bottom of the bed and the conductivity cell in order to avoid distortion of the signal read by the cell. A typical record of outlet tracer concentration is shown in Figure 3 and, from the sensitivity of the curves representing Eq. 6, to the value of Pe_L, it may be guessed that the values of D_L obtained are accurate to within $\pm 15\%$.

The large majority of our experiments were performed with beds of closely sized glass ballotini (625 μm average particle diameter) packed inside this test column, but a significant number of experiments were initially performed with other test columns, at ambient temperature, in order to test the validity of the experimental approach. Ballotini (462 μm) and closely sized sand (496 μm) were used (separately) to pack both a tube 0.75 m long with a 34 mm ID and a tube 2.025 m long with a 32 mm ID; packed beds of glass spheres (either 3.0 mm or 3.5 mm in diameter) were also used, inside an acrylic tube 2.00 m long with a 100 mm I.D. The porosity of the various packings was always between 0.37 and 0.40.

Results and Discussion

The preliminary experiments were devised to test the experimental method and check the relative importance of column dimensions, particle size, and particle shape. The results obtained are shown in Figures 5, 6 and 7. Figure 5 shows that the values of D_L are insensitive to column size for the sizes tested, while Figure 6 confirms the expected insensitivity of D_L to changes in particle size. The results confirm that flow distribution must be very nearly uniform in all packed beds, as might be expected, considering that D/d was always higher than 30, and in most experiments $D/d > 75$ (see Vortmeyer and Schuster (1983), for wall effects). Figure 7 shows that particle shape is a significant parameter, with higher values of D_L (that is, lower Pe_L) being observed in beds of sand. A similar finding has been reported by Han et al. (1985), who also report that a spread in the size distribution of the particles making up the packing resulted in higher values of D_L.

The results of our experiments with glass ballotini beds at different temperatures (all in the 3.00 m long tube) are represented in two alternative plots in Figures 8 and 9. Both plots show the dependence of Pe_L on Sc, and it may be seen that

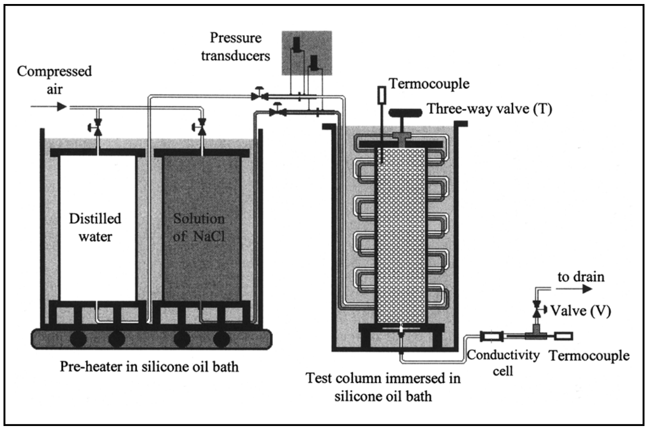

Figure 4. Experimental setup.

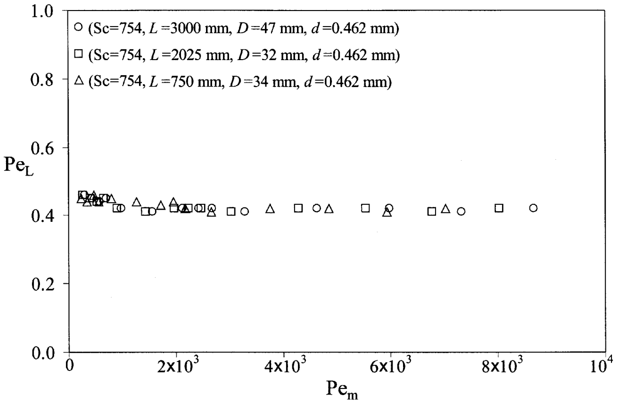

Figure 5. Effect of bed length on D_L (beds of glass ballotini).

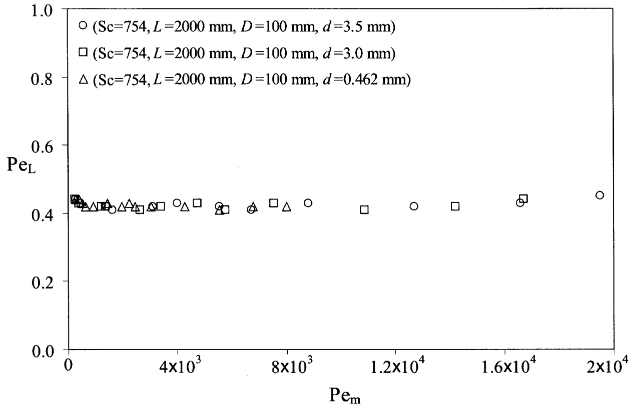

Figure 6. Effect of particle size on D_L (beds of glass spheres or ballotini).

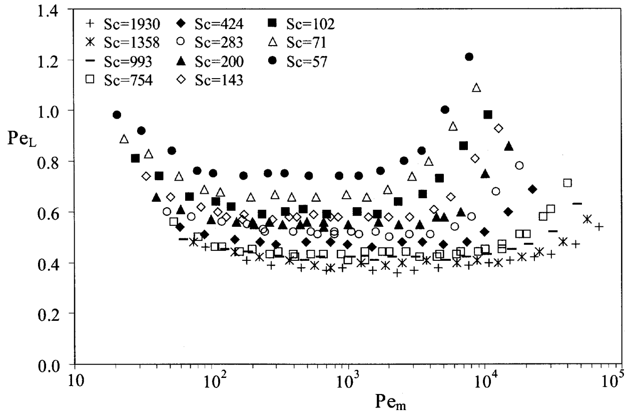

Figure 8. Dependence of Pe_L on Pe_m for different values of Sc (glass ballotini).

the dependence is slight for the higher values of Sc (say for Sc of order 750 and above). A similar finding has been reported by Delgado and Guedes de Carvalho (2001), with regard to the influence of Sc on transverse dispersion.

At the lower end of the range of Pe_m investigated (say for $Pe_m < 20$), there seems to be a tendency for Pe_L to become independent of Sc, even if the values of D_L are still significantly above D_m. In the intermediate range, $100 < Pe_m <$ 5,000, values of Pe_L are very nearly constant, for each value of Sc. Finally, Pe_L is seen to increase with Pe_m for high values of this variable, and it may be seen that, for $Re > 25$, Pe_L only depends on Re, presumably because diffusional effects are totally masked by the turbulence in the interstices of the bed. However, it seems an acceptable guess to expect that only for $Re > 100$ will diffusional effects become completely masked, when values of Sc get closer to 1.

Some workers have measured axial dispersion for the flow of supercritical carbon dioxide through fixed beds and this provides important new data in the range $1.5 < Sc < 20$. However, the various authors fail to recognize the direct dependence of Pe_L on Sc. Catchpole et al. (1996) represent their data and those of Tan and Liou (1989) in a single plot (their Figure 4) of Pe_L vs. Re. The majority of points are in the range $1 < Re < 30$, and the data of both groups, together, define a horizontal cloud with midline at about $Pe_L \cong 0.8$, spreading over the approximate range $0.3 < Pe_L < 1.1$. Although the direct effect of Sc cannot be brought into evidence in that plot, it can be said that there appears to be agreement, in broad terms, with what could be expected from our data in Figure 9.

The data of Yu et al. (1999), again for the flow of supercritical carbon dioxide, are for $0.01 < Re < 2$ and $2 < Sc < 9$. Here, it is worth noting that the modeling work of Coelho et al. (1997) gives theoretical support to experimental findings for low Re, both for spherical and nonspherical particles. No influence of Sc on Pe_L is detected, but this is what might be expected, considering that the values of Re are generally very low, with the consequence that the points are partly in the range where dispersion is totally determined by molecular diffusion. Their values of Pe_L at the higher values of Pe_m seem to be much too low; this may be because the particles used in most experiments are too small (below 0.15 mm) and this is known to yield enhanced dispersion coefficients, possibly due to particle agglomeration [see Gunn (1987), Perkins and Johnston (1963), and Hiby (1962)].

It is worth comparing the data now made available with the predictions of well-known correlations. In Figure 10, the lines corresponding to the correlations of Gunn (1969) and of Tsotsas and Schlunder (1998) are represented for the higher

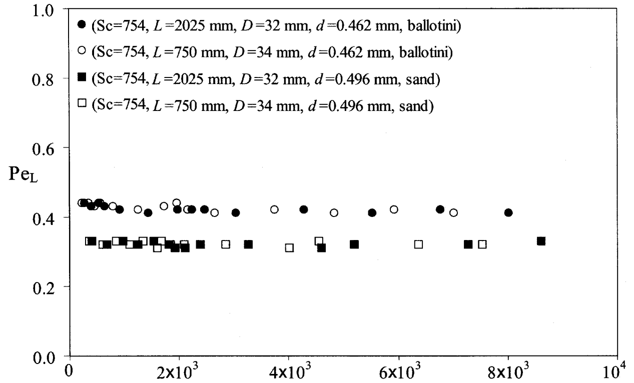

Figure 7. Effect of particle shape on D_L.

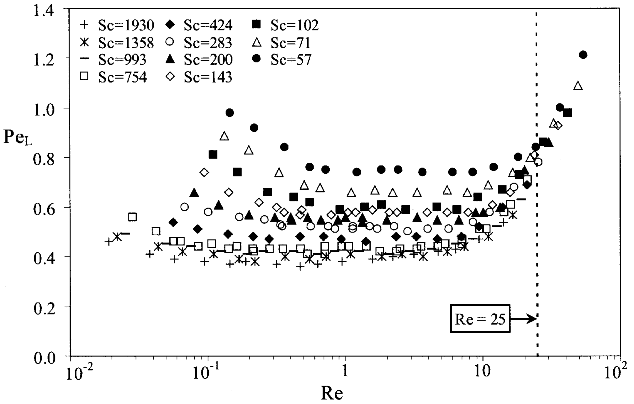

Figure 9. Dependence of Pe_L on Re for different values of Sc (glass ballotini).

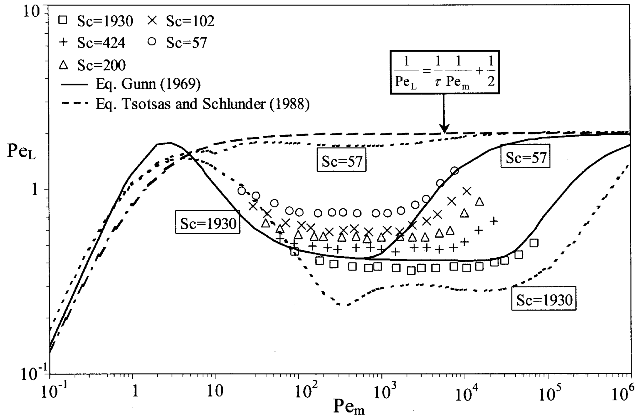

Figure 10. Comparison between experimental data and correlations.

and lower values of Sc in our experiments. It can be seen that the correlation of Gunn (1969) is not sensitive to changes in Sc, for $Pe_m < 10^3$ and the correlation of Tsotsas and Schlunder (1988) is much too sensitive to variations in Sc.

Conclusions

The present work increases our knowledge about dispersion in packed beds by providing important new data on the effect of fluid properties on the value of the axial dispersion coefficient.

The data reported cover the approxiamte ranges $20 < Pe_m < 10^5$ and $57 < Sc < 1{,}930$, and the results show that there is a near independence between Pe_L and Sc for values of the latter parameter above about 700. For lower values of Sc, down to about 50 (the lowest in the present study), Pe_L is seen to increase with a decrease in Sc, but "gas behavior" is still far from being reached.

For Re above about 25, Pe_L is seen to depend only on Re (and not on Sc), as might be expected. The data were used to test available correlations for the prediction of D_L, and it is shown that the well-known expressions of Gunn (1969) and Tsotsas and Schlunder (1988) are rather inaccurate in the region $10^2 < Pe_m < 2\times10^3$.

Notation

C = solute concentration
C_0 = initial concentration of solute
C_s = inlet concentration of solute, after step
d = diameter of inert particles
D = diameter of packed bed
D_L = longitudinal dispersion coefficient
D_m = molecular diffusion coefficient
D'_m = effective molecular diffusion coefficient ($= D_m/\tau$)
$F = (C - C_0)/(C_S - C_0)$
$Pe_L(\infty)$ = asymptotic value of Pe_L when $Re \to \infty$
t = time
$\bar{t}$ = mean residence time
u = interstitial velocity
z = coordinate along tube

Greek letters

ϵ = bed voidage
μ = dynamic viscosity
θ = dimensionless time
ρ = density
τ = tortuosity

Dimensionless groups

Pe_L = Peclet number based on dispersion coefficient ($= ud/D_L$)
Pe_m = Peclet nember based on molecular diffusion coefficient ($= ud/D_m$)
Re = Reynolds number ($= \rho ud/\mu$)
Sc = Schmidt number ($= \mu/\rho D_m$)

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