Lateral Dispersion in Liquid Flow Through Packed Beds at $Pe_m < 1{,}400$

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Introduction

The problem of solute dispersion in underground water movement has attracted interest from the early days of this century (Slichter, 1905), but it was only since the 1950s that the general topic of hydrodynamic dispersion, or miscible displacement, became the subject of more systematic study. This topic has interested hydrologists, geophysicists, petroleum and chemical engineers, among others, and for some time now it has been treated at length in books on flow through porous media (such as Bear, 1972; Scheidegger, 1974).

Some books on chemical reaction engineering (such as Wen and Fan, 1975; Carberry, 1976) treat the topics of ''axial'' and ''radial'' dispersion in detail, and it is generally observed that data for liquids and gases do not overlap, even in the ''appropriate'' dimensionless representation. The reason for this is suspected to be that the Schmidt number (Sc) is of order unity for most gas mixtures, whereas for solutes in water at room temperature, Sc is typically of order 500−1,500. Curiously, there seem to be no data available for the intermediate range $2 < Sc < 500$.

Also, available values of D_T for water flow through packed beds show significant scatter, and it is not clear whether this is associated with the experimental techniques used, or with the variability of such parameters as particle shape, particle-size distribution, and the exact value of Sc at which the experiments were conducted. The present work was undertaken to clarify these matters, since accurate values of D_T were required in a parallel study of the dissolution of large buried spheres, at Peclet number, $Pe_m < 600$ (Guedes de Carvalho and Delgado, 1999).

Measurements of longitudinal and lateral dispersion are normally carried out separately, and it is generally recognized that ''experiments on lateral dispersion are much more difficult to perform than those on longitudinal dispersion'' (Scheidegger, 1974, p. 304). The more widely used techniques for the measurement of lateral dispersion are the continuous source method and the instantaneous finite source method (such as Gunn, 1968), which rely on the injection of a tracer in the flowing liquid, followed by tracer detection at several points downstream of the injection point.

In recent years, nuclear magnetic resonance has been used to determine both diffusion and dispersion coefficients (such as Gibbs et al., 1992 and Baumeister et al., 1995) with significant advantages, but the method seems to be limited to very low fluid velocities.

An alternative technique for measuring transverse dispersion was developed by Coelho and Guedes de Carvalho (1988a) in a first step toward understanding mass transfer between the surface of large bodies that are buried in granular beds and the fluid flowing around them (Coelho and Guedes de Carvalho, 1988b; Burmester et al., 1990; Guedes de Carvalho et al., 1991). That technique was mostly used to study transverse dispersion in gas flow, but some 20 experimental values of D_T were reported in the original article for water flow through beds of glass ''ballotini.'' The technique is simple to use, and after outlining the theory behind it, we report the data obtained on the influence of particle size and size distribution, particle shape and liquid properties (through changes in temperature of operation) on the coefficient of transverse dispersion. Our study is limited to $Pe_m < 1{,}400$ by virtue of the technique used.

Measurements of transverse dispersion have been reported in the literature for liquid flow through beds of sand, of spheres, and of ion-exchange resin. In our literature survey we found (for the range $Pe_m < 1{,}400$) some 70 data points for the transverse dispersion coefficient (D_T) in beds of spheres with sizes between 74 μm and 1.5 mm, and 75 data points for D_T in beds of sand with sizes between 53 μm and 750 μm, always for $Sc \geq 500$. In the present note we report some 230 new experimental values of D_T for water flow through beds of silica sand of between 200 μm and 500 μm average particle size, with both narrow- and wide-size distributions and for $140 \leq Sc \leq 1{,}200$.

No attempt is made at presenting a mechanistic interpretation of the data obtained at this stage, since we were limited to $Pe_m < 1{,}400$.

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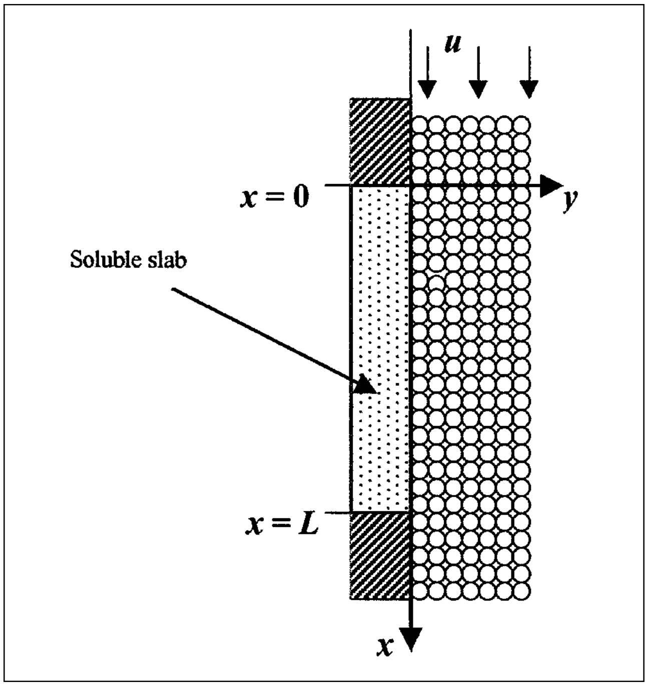


Figure 1. Flow through packed bed near soluble surface.

Outline of the Method

Mass transfer from a flat surface aligned with the flow

Figure 1 shows a section through a packed bed along with liquid is flowing close to a flat wall, part of which ($0 < x < L$) is slightly soluble. Liquid flow will be taken to be steady, with uniform average interstitial velocity u, and if the concentration of solute in the liquid fed to the bed is C_0 and the solubility of the solid wall is C^*, a mass-transfer boundary layer will develop, across which the solute concentration drops from $C = C^*$, at $y = 0$, to $C \rightarrow C_0$, for large y.

The question of how large is meant by a ''large y'' needs some clarification. Obviously, if L were only of the order of a few particle diameters, and u were large, the concentration of solute would fall to C_0 over a distance of less than one particle diameter. In that case, flow in the bulk of the packed bed would have little influence on the mass-transfer process, which would be dominated by diffusion in a thin layer of liquid adjacent to the soluble surface. Already for large L and low u, the thickness of the mass-transfer boundary layer grows from zero, at $x = 0$, to a value of several particle diameters, at $x = L$, and the process of mass transfer can then be determined by competition between advection and dispersion in the bulk of the bed. Now, it is well known (such as Vortmeyer and Schuster, 1983) that the voidage of a packed bed (and therefore the fluid velocity) is higher near a flat containing wall, but in the case of our experiments it can be considered that such a nonuniformity will have negligible effect. For one thing, we work with bed particles of between 0.2 and 0.5 mm, and therefore the region of increased voidage will be very thin. Furthermore, because the inert particles making up the bed indent the soluble surface slightly, as dissolution takes place (and this slight indentation is easily confirmed when the piece of soluble solid is removed from within the bed) there is in fact virtually no near wall region of higher voidage. Confirmation of these assumptions is given by the results of the experiments described below.

If we restrict our analysis to those situations for which the mass-transfer boundary layer extends over several particle diameters, and if a small control volume is considered, inside this boundary layer, with side lengths δx, δy and unity (perpendicular to the plane of the figure), a steady-state material balance on the solute leads to

$$u\frac{\partial C}{\partial x} = D_T\frac{\partial^2 C}{\partial y^2} + D_L\frac{\partial^2 C}{\partial x^2}, \qquad (1)$$

where D_T and D_L are the dispersion coefficients in the cross-stream and longitudinal directions, respectively. If the boundary layer is thin compared to the length of the soluble slab, the last term on the righthand side of Eq. 1 is likely to be negligible, since the surface $y = 0$, $0 < x < L$ is a surface of constant concentration ($C = C^*$).

As detailed by Coelho and Guedes de Carvalho (1988a), a conservative criterion for this approximation to be valid is $L/d > 20$, where d is the average size of the particles in the bed; Eq. 1 then reduces to the equation of diffusion in one dimension:

$$u\frac{\partial C}{\partial x} = D_T\frac{\partial^2 C}{\partial y^2}, \qquad (2)$$

to be solved with

$$C = C_0 \qquad x = 0 \qquad y > 0 \qquad (3a)$$

$$C = C^* \qquad x > 0 \qquad y = 0 \qquad (3b)$$

$$C \rightarrow C_0 \qquad x > 0 \qquad y \rightarrow \infty. \qquad (3c)$$

From the solution

$$\frac{C - C_0}{C^* - C_0} = erfc\left(\frac{y}{2\sqrt{D_T x/u}}\right) \qquad (4)$$

it is possible to obtain the instant rate of the solid dissolution over the whole-slab surface

$$n = \int_0^L NZ\,dx = (C^* - C_0)\epsilon ZL\left(\frac{4D_T}{\pi L/u}\right)^{1/2}, \qquad (5)$$

where $N = -D_T\epsilon(\partial C/\partial y)_{y=0}$ and Z is the slab width.

(Note that there is an initial length of slab over which this solution is not valid, but for a long enough slab the contribution of this ''entry'' length is negligible.)

If we define an average mass-transfer coefficient $k = n/A(C^* - C_0)$, where A is the exposed area of the solid, we get

$$k = \epsilon\left(\frac{4D_T}{\pi L/u}\right)^{1/2}, \qquad (6)$$

and this shows how the measurement of the rate of dissolution of the solid can be used to determine the coefficient of transverse dispersion in the bed.

A simple way of checking the result in Eq. 6 is given by the predicted proportionality between k and the inverse square root of L. Experiments performed by Coelho and Guedes de Carvalho (1988a) with a wide range of slab lengths, both for the dissolution of benzoic acid in water and the sublimation of naphthalene in air, confirm the general validity of the preceding theory, provided the approximate criterion

$$\frac{L}{d} \geq 0.62\left(\frac{ud}{D_m}\right) \tag{7}$$

is observed, where D_m is the molecular diffusion coefficient of the solute. When this criterion is not observed, the near-wall film resistance to diffusion has to be taken into account, and approximate ways of doing that are described by Coelho and Guedes de Carvalho (1988a).

Mass transfer from a cylinder aligned with the flow

For practical reasons, it proves simpler to perform experiments in which the dissolving solid is a cylinder aligned with the flow direction, and it is important to know the theoretical expressions relating the average mass-transfer coefficient with the coefficient of dispersion, D_T, for that situation.

Fortunately, under appropriate conditions that are easy to reproduce in the laboratory, the thickness of the mass-transfer boundary layer is small compared to the radius of the dissolving cylinder, and under such circumstances the analysis for dissolution from a flat surface just presented is still applicable with good accuracy.

However, there are instances in which this simplification is not valid, and an exact solution can be worked out in cylindrical coordinates, as shown by Coelho and Guedes de Carvalho (1988a).

The resulting expression for k is cumbersome to evaluate, but for small values of the parameter $\theta_c = D_T t_c / a^2$, where $t_c = L/u$ is the time of contact between liquid and solid, a good approximation is

$$k = \epsilon\left(\frac{4D_T}{\pi t_c}\right)^{1/2}\left(1 + \frac{\sqrt{\pi}}{4}\theta_c^{1/2} - \frac{1}{12}\theta_c + \frac{\sqrt{\pi}}{32}\theta_c^{3/2} - \ldots\right). \tag{8}$$

If we make $Y = 1 + \theta_c^{1/2}\sqrt{\pi}/4 - \theta_c/12 + \theta_c^{3/2}\sqrt{\pi}/32$, it is instructive to plot Y vs. θ_c, as shown in Figure 2, and from the plot it becomes clear that Eq. 6 gives an accuracy better than 5% if $\theta_c < 0.01$, which is a condition met in most experiments of practical interest. For values of θ_c up to $\theta_c = 0.4$, the four terms in Y can be used instead of the infinite series on the righthand side of Eq. 8, with an error of less than 1% in k.

Experiment

In each experiment, a cylinder of a slightly soluble solid, buried in a packed bed, in alignment with the flow direction, is allowed to dissolve gradually in a stream of liquid (typically water) that is forced through the packing at a constant flow rate. The rate of dissolution is obtained by measuring the liquid flow rate and solute concentration at the bed outlet (C_{out}).

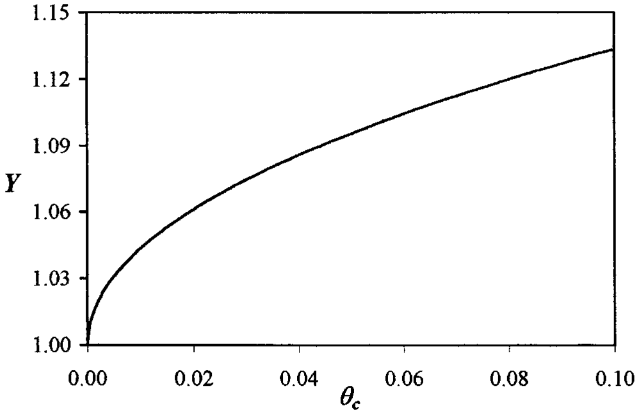


Figure 2. Dependence of Y on θ_c.

Experiments were performed on the dissolution of cylinders of benzoic acid buried in beds of sand in the same type of rig as that used by Coelho and Guedes de Carvalho (1988a). The cylinders of benzoic acid were 20 mm in diameter and their lengths were either 85, 155, 215 or 250 mm. The temperature of the packed bed could be adjusted over a narrow range, and experiments were performed at 293, 298, 303 and 308 K.

The silica sand used in the experiments was initially washed, dried, and sieved in narrow cuts, and one "mixed lot" was also prepared by blending two batches of different sizes. The size distributions of all the batches used in the experiments were determined by a laser diffraction technique and are given in Table 1. The tortuosity of the packed beds was taken to be $\tau = 1.4$, the value recommended by most authors for random packings of closely sized particles and confirmed by our results, reported below.

Results and Discussion

Having worked with five batches of sand and typically with two cylinder lengths (sometimes three) for each batch, we made over 200 determinations of D_T, not counting the repeated runs. A complete tabulation of the data points will be supplied to any interested reader on request.

The reproducibility of the experiments was tested by independently repeating the measurement of k under identical

Table 1. Characteristics of the Sand Beds

				Particle Diameter ($< \mu$m)									
Lot	d mm	ϵ	τ	10% Vol.	20% Vol.	30% Vol.	40% Vol.	50% Vol.	60% Vol.	70% Vol.	80% Vol.	90% Vol.	100% Vol.
A	0.219	0.40	1.4	161	180	195	210	225	241	259	281	313	476
B	0.297	0.35	1.4	185	224	250	274	296	320	346	378	422	625
C	0.322	0.42	1.4	236	261	281	300	319	339	360	388	426	625
D*	0.409	0.32	1.4	221	267	305	342	380	424	476	549	665	900
E	0.496	0.33	1.4	277	340	389	434	480	530	588	660	756	900

*Lot D is a mixture 50% w/w of lots B and E.

operating conditions for a considerable number of data points. In the vast majority of cases, repeated measurements of k did not differ by more than 5%, which, in the light of Eq. 6, suggests that the values of D_T reported below are reproducible to within less than 10%.

An important aspect in the experimental results is that the outlet concentration of solute, C_{out}, was constant over very long periods in each experiment. Considering that the surface of the soluble cylinders was initially very smooth and that it was completely covered with slight indentations (from the inert particles) at the end of the run, we can conclude that any "near-wall effect" has a negligible influence on the process of mass transfer.

In the analysis of the data collected, an important aspect to be considered, as a check on the experimental method, is the influence of the length of the test cylinder (L) on the measured value of D_T. In reality, the two variables are independent provided the criterion given by Eq. 7 is satisfied, as required, and the values of D_T plotted in Figure 3 illustrate this independence.

Figure 4 gives a direct measure of the effect of temperature on D_T and also, from a comparison between the circles and the squares, it can be concluded that an increase in particle size determines an increase in D_T for any given u, the effect being more pronounced at the higher flow rates.

In order to understand the influence of fluid velocity on the dispersion coefficient, it is important to consider the limiting case where $u \rightarrow 0$. The effective diffusion coefficient, based on the whole cross-sectional area, is then given by $D_m(\epsilon/\tau)$, where D_m is the coefficient of molecular diffusion and τ is the tortuosity of the packed bed with regard to diffusion. Now, because the definition of D_T is based on the open area of the packing, $D_T \rightarrow D'_m = D_m/\tau$, in the limit $u \rightarrow 0$. In the plots of Figure 3 we indicate the values of D'_m obtained with $\tau = 1.4$ on the vertical axis, and alignment with the experimental points is good.

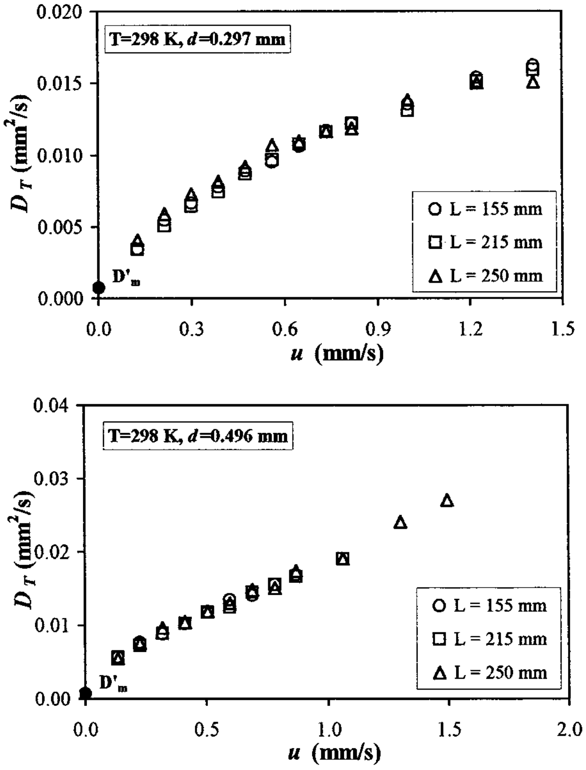


Figure 3. Variation of D_T with u for different lengths of test cylinder.

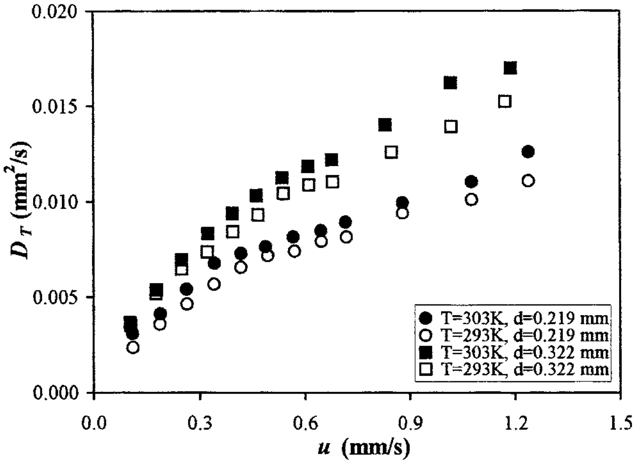


Figure 4. Effect of temperature on variation of D_T with u.

As the velocity of the fluid is increased, the contribution of convective dispersion tends to become dominant over that of molecular diffusion. According to several authors (see Hiby, 1962; Wilhelm, 1962; Gunn, 1968; Bischoff, 1969; Coelho and Guedes de Carvalho, 1988a), $D_T \rightarrow ud/K$ for high enough values of u, with $K \cong 12$ for beds of closely sized particles. If it is assumed that the diffusive and convective components of dispersion are additive, then $D_T = D'_m + ud/K$, and this may be written in dimensionless form as

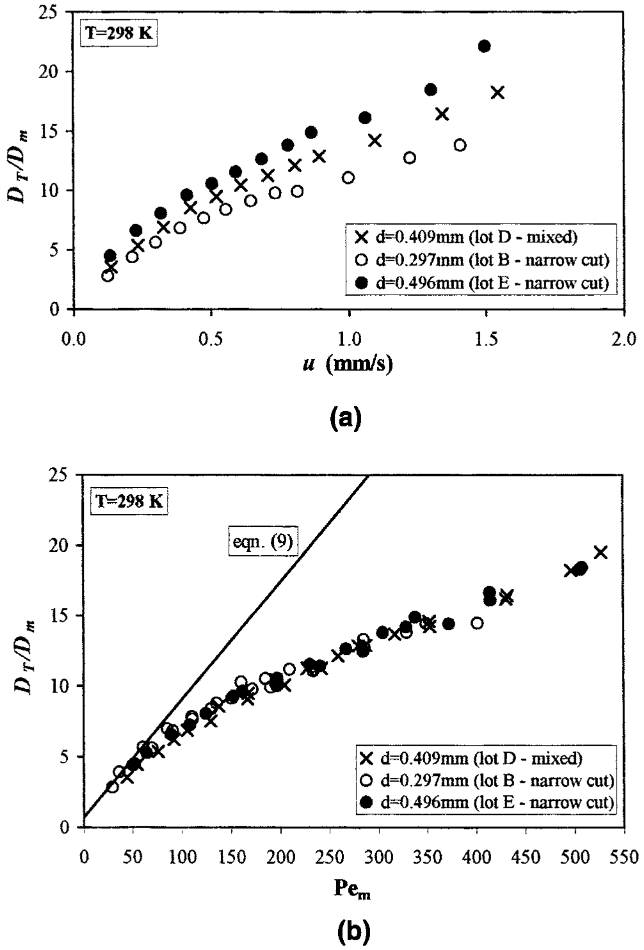
$$\frac{D_T}{D_m} = \frac{1}{\tau} + \frac{1}{12}\frac{ud}{D_m}. \tag{9}$$

This equation is known (such as Coelho and Guedes de Carvalho, 1988a) to give a fairly accurate prediction of D_T for gas flow through packed beds, and it also has the correct asymptotic behavior for liquids, both for low and high Pe. However, over an intermediate range of values of ud/D_m, Eq. 9 is not appropriate for the description of dispersion in liquids, as pointed out by several of the authors mentioned earlier.

Fortunately, data on dispersion in randomly packed beds of near-spherical particles that are closely sized lend themselves to simple correlation by means of dimensional analysis. Indeed, it is reasonable to assume the functional dependence

$$D_T = \phi(u, d, \rho, \mu, D_m) \tag{10}$$

for beds of a given porosity and tortuosity, where ρ and μ are the density and viscosity of the fluid, respectively. Using



(a)

(b)

Figure 5. (a) Effect of particle mixture on variation of D_T with u; (b) variation of D_T/D_m with Pe_m for lots B, D and E.

Buckingham's Π theorem, it can then be concluded that

$$\frac{D_T}{D_m} = \Phi\left(\frac{ud}{D_m}, \frac{\mu}{\rho D_m}\right), \qquad (11)$$

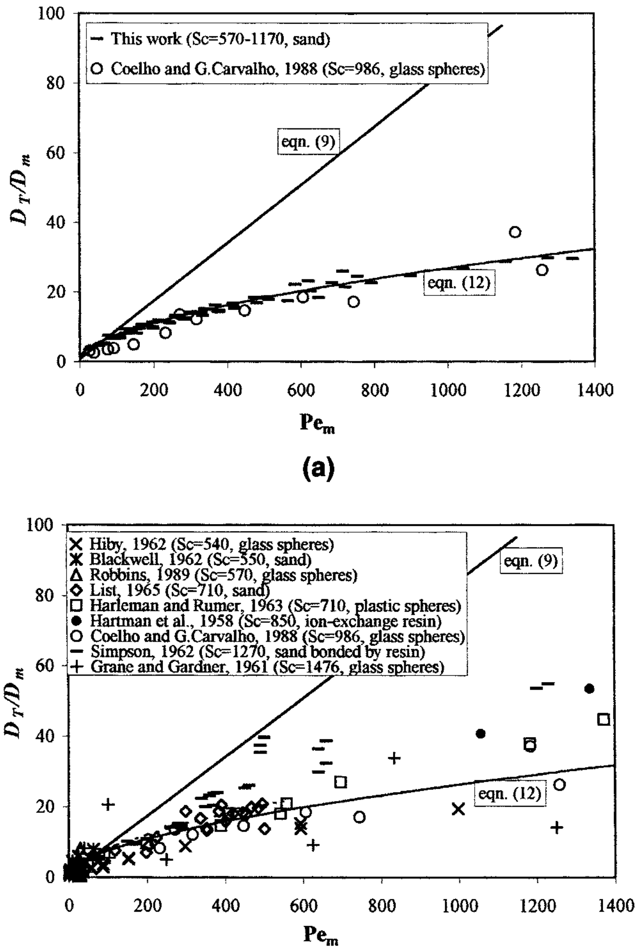
where the dimensionless groups $Pe_m = ud/D_m$ and $Sc = \mu/\rho D_m$ are easily identified, and Eq. 11 suggests that the experimental data be plotted as (D_T/D_m) vs. Pe_m. Equation 11 is expected to hold for beds of (nearly) monosized particles, and it is interesting also to test its applicability to beds with a wide distribution of particle sizes. This is done in Figure 5, where the average particle size was used in the calculation of Pe_m. It can be seen that the "spread" of raw data in the plot of (D_T/D_m) vs. u is virtually eliminated in the plot of (D_T/D_m) vs. Pe_m, thus suggesting that Eq. 11 can be applied to beds with a wide distribution of particle sizes, if the average diameter is considered.

In Figure 6a we show the experimental points obtained with the present technique, including those of Coelho and Guedes de Carvalho (1988a) and Delgado and Guedes de Carvalho (2000), together with the fitted line

$$\frac{D_T}{D_m} = \frac{1}{\tau} + 0.50\,Pe_m^{0.57}, \qquad (12)$$

which is seen to represent the data very well for $Pe_m > 80$. Below this value of Pe_m, Eq. 9 should be used instead. The points for "glass ballotini" are very close to those for sand and the conclusion seems to be that particle shape has only a small influence on lateral dispersion (for random packings of "isometric" particles).

The data in Figure 6a were obtained at five distinct values of Sc in the range $550 < Sc < 1200$, and the fact that they all fall along a single line is in agreement with the finding of Blackwell (1962) that D_T/D_m is independent of Sc, for $Sc \geq 500$. Blackwell covered the $500 \leq Sc \leq 3.15 \times 10^5$ range, using water and solutions of glycerol at ambient temperature. Now, since all the published values of D_T for liquids refer to $Sc \geq 500$, they would be expected to fall close to Eq. 12, for $Pe_m > 80$.



(a)

(b)

Figure 6. Comparison of data for Sc > 550.

(a) Present method; (b) other data.

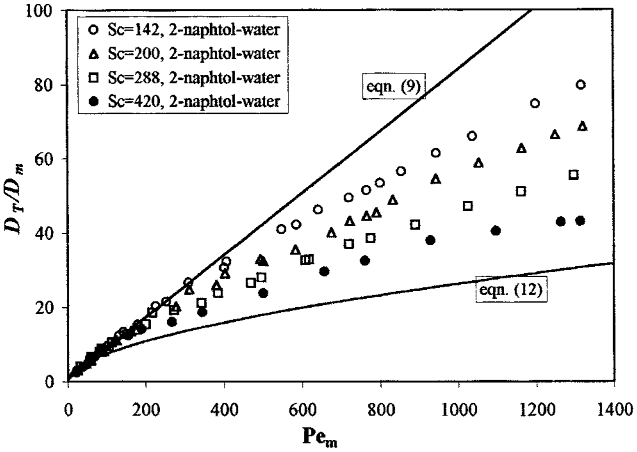


Figure 7. Effect of *Sc* on D_T/D_m.

The comparison is made in Figure 6b, where significant scatter about Eq. 12 is obvious in the data of Grane and Gardner (1961). Also, two points of Harleman and Rumer (1963) and two points of Hartman et al. (1958) are some 30 to 50% above Eq. 12. Many of the points reported by Simpson (1962) seem to fall outside of the general trend, but it is to be noted that they were obtained in beds of sand bonded by resin, and this leads to the formation of particle agglomerates, with a resulting increase in values of D_T. Blackwell (1962) had already observed that in a bed with sand particles of 74 μm to 840 μm, values of D_T were considerably higher than expected as a result of particle agglomeration (which gives a larger "apparent particle diameter"). The same phenomenon is probably also the explanation for the high values of D_T obtained by Van der Poel (1961), who worked with particles sizes of around 0.111 μm.

At the lower end of Pe_m, only the points of Hiby (1962) seem to fall consistently below Eq. 12; however, it is not to be excluded that Hiby's data (at low Pe_m) are subject to wall effect, for the width of the test channel (4 mm) was only eight times the diameter of the particles. Experimental values of D_T for $Sc < 500$ with liquids were only recently determined (Delgado and Guedes de Carvalho, 2000) using the experimental technique described earlier to study dispersion at temperatures of up to 70°C (using 2-naphthol instead of benzoic acid). Values of D_T were determined for Sc down to 142, and the results are shown in Figure 7, where it is clearly seen that D_T/D_m is strongly dependent on Sc, for $Sc < 500$. This is a new finding, and it shows how the gap between gas behavior and liquid behavior is bridged for $Sc < 500$.

Conclusions

Our work shows that values of D_T/D_m in packed beds are independent of Sc in the range $Sc > 500$, but strongly dependent on Sc for $140 < Sc < 500$. This is an important first step toward understanding the difference between gas and liquid behavior when dispersion is involved. The difference in behavior between beds of spheres and beds of sand resulting from the difference in particle shape is shown to be slight, and it is reported that dispersion in a bed with a mixture of particle sizes follows the prediction for beds of nearly mono-sized particles if the average particle size is used in the calculation of Pe_m.

It is suggested that Eq. 9 gives a good prediction of D_T for $Pe_m < 80$, for all Sc, whereas Eq. 12 should be used for $80 < Pe_m < 1{,}400$ and $Sc > 500$.

Notation

a = radius of soluble cylinder, m
D'_m = apparent molecular diffusion coefficient ($= D_m/\tau$), m^2/s
K = asymptotic value of ud/D_T when $Re \to \infty$
L = length of solid slab or cylinder, m
N = local flux of solute, kg/m^{2}s
n = total mass transfer rate, kg/s
r = radial coordinate, m
x = Cartesian coordinate, m
y = Cartesian coordinate, m
Y = sum of first four terms in series on righthand side of Eq. 8
ϵ = bed voidage
θ_c = dimensionless time of contact

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