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Diameter-Dependent Dispersion in Cylindrical Bead Packs

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It has recently been argued that longitudinal dispersion in cylindrical packed beds is enhanced due to radial heterogeneities in the porosity and resulting velocity field. In particular, it is predicted that the time scale to attain asymptotic dispersion in a packed bed of radius R is proportional to R^2/D_T, where D_T is the transverse dispersion constant in a homogeneous beadpack, and that the asymptotic longitudinal dispersion constant increases with R/d, where d is the bead diameter. We have measured the asymptotic dispersion rates for packed beds with $R/d = 9.5$, 12.7, and 25.4 and find good qualitative agreement with the predicted dependence on R/d. *AIChE J,* 54: 2024–2028, 2008
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Introduction

Conventional wisdom has been that the asymptotic hydrodynamic dispersion coefficient in a cylindrical beadpack depends on the ratio of the cylinder radius (R) to the bead diameter (d), R/d, only for small values of this ratio. For example, Gunn[1] has speculated that for $R/d > 5$ the longitudinal dispersion coefficient will not depend on the cylinder diameter. A recent theoretical analysis and simulation study,[2,3] however, concluded that this is not the case, and that the asymptotic dispersion coefficient increases with R/d, even for large values of this ratio.

In this article, we present new data for the longitudinal dispersion coefficient in bead packs of sufficient length to assure that the dispersion is fully developed (asymptotic). The results support the predictions of Refs. 2 and 3 that for a given particle Peclet number, $Pe = \bar{v}d/D_m$, where $\bar{v}$ is the mean flow velocity and D_m is the molecular diffusivity, the value of the asymptotic longitudinal dispersion constant exhibits a strong dependence on the ratio R/d.

It is generally accepted that the longitudinal dispersion coefficient, D_L, depends on length of a packed bed. Han et al.[4] published a careful and extensive study of this subject. On the other hand, Gunn's speculation and the neglect of

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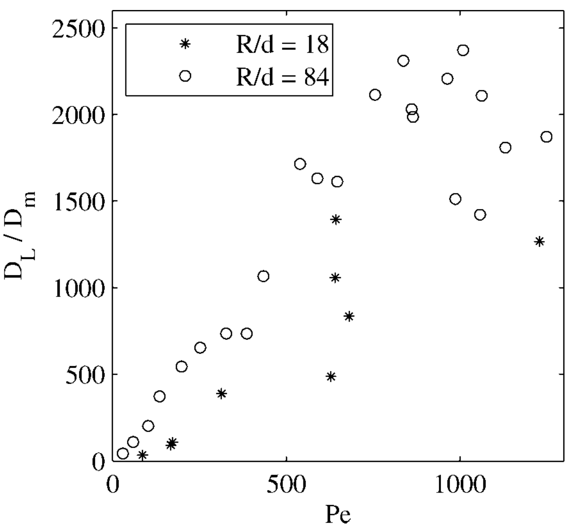

Figure 1. Longitudinal dispersion as a function of the Peclet number, *Pe*, in a packed column of glass spheres.[6]

this issue in Delgado's[5] recent extensive review of dispersion experiments indicate that the dependence on the column radius to bead diameter ratio is not felt to be important. There is, however, some earlier evidence supporting the dependence of D_L on R/d. For example, the well-known results of Pfannkuch[6] suggest that the dispersion coefficient in a column packed with glass spheres does indeed depend on the ratio R/d (Figure 1).

Additional evidence for the effect of R/d on dispersion comes from chromatographic theory and experiment. Knox and Parcher,[7] for example, found dispersion (plate height, in their nomenclature) to increase with R/d for the range studied ($R/d < 11.5$). They also show that the dispersion exhibits a much weaker dependence on Peclet number (reduced velocity in their nomenclature) in an infinite column than in a walled column. (An infinite column is one in which "a sample injected centrally at the head of a column fails to reach the disturbed packing region close to the walls by the time the sample reaches the foot of the column.").

One point of view is that, to attain asymptotic dispersion, the column must be sufficiently long that solute can diffuse over an area proportional to d^2. While this criterion might be appropriate for homogeneous packed beds, it fails to account for the heterogeneities of packing and flow near the cylinder's wall. In Ref. 2 it was argued that for cylindrical beadpacks, R^2/D_T is the appropriate time for longitudinal dispersion to be fully developed, where D_T is the transverse dispersion coefficient in an unconfined system.

Experimental Setup and Methods

In the experiments reported here, we used plastic tubes with inner diameters of 3/8, 1/2, and 1 in., respectively. We packed them with glass beads 500 μm in diameter (Q-Beads by Quackenbush Company, Inc.). We initially placed dry beads within the tubes using gravity and agitation supplied by tapping the tubes' exterior to aid in the packing process. Improvements in packing were achieved by means of further agitation in conjunction with water flow through the bead pack. Porosity estimates were made by evacuating the packing from each tube and measuring the collected bead mass to determine each tube's packing volume. This method approximated a fractional porosity of 0.43, 0.40, and 0.41 for the tubes of 3/8, 1/2, and 1 in. diameter, respectively.

To measure hydrodynamic dispersivity, a pulse of potassium chloride is injected in the bead pack and the conductivity is measured at a downstream port as a function of time. The diffusivity of potassium chloride in dilute solutions is $D_m = 1.85 \times 10^{-5}$ cm^2/s.[8] The flow is controlled by a Fluid Metering Inc. QG150 pump delivering tap water at the upstream end. The conductivity is measured by a Hewlett Packard 4277A LCZ Meter at the downstream port. The experimental system is illustrated in Figure 2.

According to theory, dispersion will be fully developed when tD_T/R^2 is of order one, where t is time, R is the tube radius and D_T is the transverse dispersion coefficient in an unconfined bead pack.[2] Fully developed dispersion is therefore predicted to be achieved for bead packs longer than $L = \bar{v}t = \bar{v}R^2/D_T$, where $\bar{v}$ is about 0.2 cm/s for the three tubes used in this study. Based on simulation data presented in Ref. 2 for a porosity of 0.4, D_T is estimated to be 2.7×10^{-4} cm^2/s. The resulting estimates for the length L required

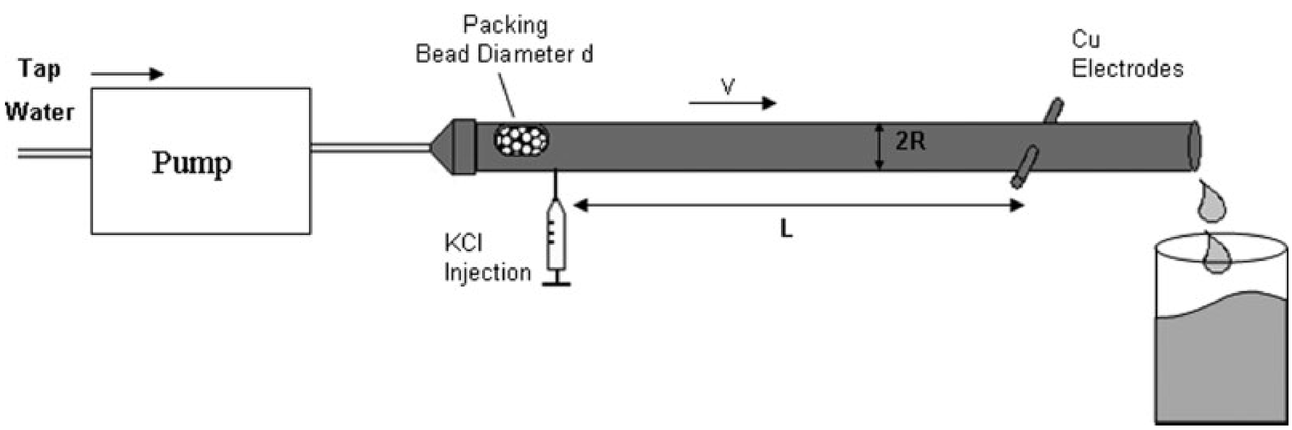

Figure 2. Experimental system.

Table 1. Experimental Column Radius and Length

$2R$ (in.)	R/d	L (ft)
3/8	9.5	4.7
1/2	12.7	8.4
1	25.4	33.5

to observe fully developed dispersion are L = 34.5 ft for the 1-in. bead pack, L = 10 ft for the 1/2-in. bead pack, and L = 5 ft for the 3/8-in. bead pack (Table 1). The experimental lengths used in the experiments reported here are shown in Table 1. Though these are somewhat shorter than our estimates, asymptotic dispersion is expected to be reached well before the length $\bar{v}R^2/D_T$ is reached (see Figure 4).

When dispersion is fully developed, the concentration profile of potassium chloride at the position x at time t is given by

$$C(x,t) = \frac{m}{\sqrt{4\pi D_L^{\mathrm{eff}} t}}\, e^{\frac{-(x-\bar{v}t)^2}{4D_L^{\mathrm{eff}} t}} \tag{1}$$

where D_L^{eff} is the effective hydrodynamic longitudinal dispersion coefficient. Correspondingly, the conductivity of the fluid at position x at time t is given by

$$\kappa = \kappa_0 + sC(x,t) \tag{2}$$

where κ_0 is the conductivity of tap water and s is the conductivity of potassium chloride per molecule. Thus, at the conductivity port located at position x_c, the dispersion coefficient is determined by a least squares fit of the data for $\kappa - \kappa_0$ to the formula

$$\frac{B}{\sqrt{4\pi D_L^{\mathrm{eff}} t}}\, e^{\frac{-(x_c-\bar{v}t)^2}{4D_L^{\mathrm{eff}} t}} \tag{3}$$

with the constraint that the time integral of this formula equals the area under the experimental curve.

Data and Analysis

Experiments were conducted on each tube to monitor the liquid stream's conductivity at a fixed position x_c as a pulse of potassium chloride passed through the bead pack. The flow rate of tap water into each tube was held at a common mean flow velocity of 0.19 cm/s. Conductivity data were collected at x_c for a set of time intervals following the injection of potassium chloride. These conductivity data were then used to determine the fitting parameters B and D_L^{eff}. The following figures present sample data sets and the corresponding fit curves for each tube (Figure 3).

Multiple data sets were collected and processed in the preceding manner for each tube. The resulting estimates for D_L^{eff} were then averaged and a standard deviation analysis was performed. The results are shown in Table 2.

To estimate the effect of packing the glass beads in the tubes, two different packings were prepared for R/d = 12.7. In the first packing, four measurements were made. For these, the mean value of D_L^{eff} was 0.0144 cm^2/s, with a standard deviation of 0.0003 cm^2/s. In the second packing, three measurements were made. For these, the mean value of D_L^{eff} was 0.0148 cm^2/s, with a standard deviation of 0.0001 cm^2/s.

Independent predictions of D_L^{eff} were obtained using the extended Aris model of axial dispersion in packed cylinders (Eq. 24, Ref. 2). We first describe this model and then discuss the resulting predictions. The extended Aris model is a generalization of Taylor-Aris dispersion to the case of a packed cylinder. The model requires knowledge of the flow profile, $\chi(r)$, in a packed cylinder and of the dispersion coefficients, D_L and D_T, in an unconfined packing of the same porous media used in the packed cylinder. The model

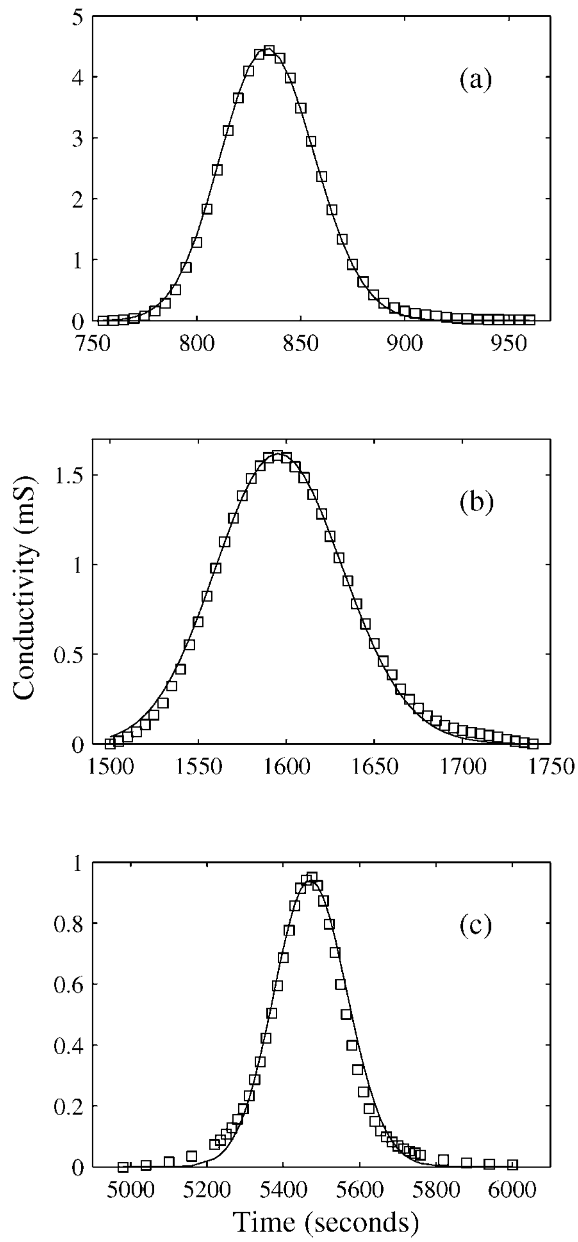

Figure 3. Conductivity as a function of time for cylinders of different radii; open squares, □, denote data, solid line denotes fitted curve.
(a) R/d= 9.5, (b) R/d = 12.7, (c) R/d = 25.4.

Table 2. Experimental Data

R/d Value	No. of Data Sets	Mean Velocity (cm/s)	Average D_L^{eff} (cm^2/s)	Std. Deviation ΔD_L^{eff}
9.5	6	0.186	0.0104	0.0002
12.7	7	0.188	0.0146	0.0003
25.4	6	0.190	0.030	0.002

Table 3. Comparison of Predicted and Experimental Data

	D_L^{eff} (cm^2/s)	
R/d	Prediction	Experiment
9.5	0.0085	0.0104
12.7	0.0114	0.0146
25.4	0.0326	0.030

describes how variations in $\chi(r)$ increase dispersion above the value of D_L; $\chi(r)$ plays a role analogous to the Poiseuille flow profile in Taylor-Aris dispersion in a tube, while D_L plays a role analogous to the molecular diffusion coefficient in the Taylor-Aris analysis.

The flow profile in a packed cylinder has been characterized in the experimental and simulation literature on packed beds.[9,10] We obtained $\chi(r)$ from 3-D pore-scale simulations of steady flow through cylinders packed with spheres. Pore velocities $v(x,y,z)$ were averaged in thin cylindrical shells to determine the profile $v(r)$, where $v = v_z$ represents the axial component of velocity, and $\chi(r) = v(r)/\bar{v} - 1$. It has been previously found that the resulting dimensionless flow profiles do not depend on the Reynolds number for $Re < 10$ (Figure 9, Ref. 2). $\chi(r)$ is roughly sinusoidal near the cylinder wall with period d, and the amplitude of these oscillations decays over a distance of $\sim 6d$. This decay distance defines the structured region of velocity, and is similar for a wide range of $R/d > 6$ (Figure 5, Ref. 2). In addition, the amplitude of the oscillations in $\chi(r)$ is essentially independent of R/d for this range of values (Figure 8, Ref. 2).

The dispersion coefficients, D_L and D_T, were obtained using 3-D pore-scale simulations of dispersion in a homogeneous packed bed of uniform spheres, using periodic boundary conditions. The dimensions of the periodic bed were 24 d $\times$ 24 d $\times$ 100 d. This homogeneous packing has a similar pore structure to the interior region of a cylindrical packed bed, where the velocity, $\chi(r)$, is essentially constant and close, but not necessarily equal, to zero. The particle Peclet number, $Pe = \bar{v}d/D_m$, used in the pore-scale simulation was similar to that used in the present experimental system. The asymptotic dispersion coefficients in the pore-scale system were $D_L/D_m = 224$ and $D_T/D_m = 14.7$ for $Pe = 476$. In order to calibrate the extended Aris model for the experimental system, these values were scaled by the factor 514/476, where $Pe = 514$ corresponds to the present experiments and $Pe = 476$ corresponds to the pore-scale simulation data. This calibration assumes D_L scales as Pe in this range. This is consistent with the literature (e.g., Ref. 11) and previous pore-scale simulation (Ref. 12). The values used to calibrate the extended Aris model were therefore $D_L/D_m = 242$ and $D_T/D_m = 15.8$. The diffusivity of potassium chloride in dilute aqueous solutions is $D_m = 1.85 \times 10^{-5}$ cm^2/s.[8]

The predictions obtained from the extended Aris model were determined using the present experimental values of R, d, and $\bar{v}$. The velocity profile $v(r)$ was obtained from pore-scale simulation of a packed cylinder with $R = 25d$, using the averaging technique described earlier. The profile $v(r)$ was then scaled to have mean value $\bar{v}$. Profiles for cylinders of smaller radius were made by truncating $v(r)$ for $r < 25 - R$, ensuring that each cylinder has a structured region with the same velocity profile (see remarks mentioned earlier on similarity of profiles for different R/d).

The extended Aris model predicts that $D_L^{eff}(t)$ approaches an asymptotic value on the time scale $t = R^2/D_T$ (Figure 4a). In the cases of $R/d = 9.5$ and $R/d = 12.7$, the predicted asymptotic dispersion coefficients are more than twice as large as that of the unconfined packing, $D_L^{eff} > 2D_L$. For $R/d = 25.4$, the predicted value is eight times greater than the

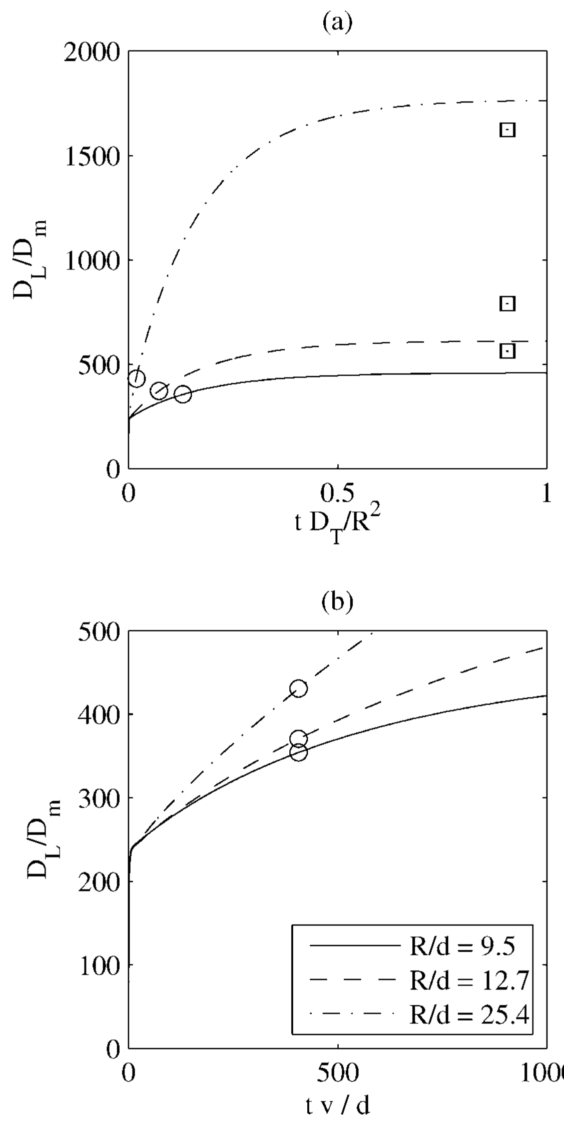

Figure 4. Theoretical predictions of dispersion for cylinders of different radius on (a) cylinder time scale and (b) convective time scale.

Circles denote predictions for a hypothetical cylinder of length $L = 406d$. Squares denote the present experimental results.

unconfined case. Note the values of $t = R^2/D_T$ are different for each cylinder radius and are significantly longer than the time for conventional breakthrough experiments, which use shorter cylinders. It is therefore interesting to evaluate the predictions for $D_L^{eff}(t)$ at shorter times, corresponding to shorter cylinders.

Figure 4b shows the predictions for $D_L^{eff}(t)$ at a fixed time $t = 406d/\bar{v}$ (corresponding to a distance eight times the largest column diameter). The predictions for $D_L^{eff}(t)$ are preasymptotic (marked in Figures 4a,b). Also, the spread in $D_L^{eff}(t)$ for three different values of R/d is relatively small in Figure 4b. If one considered only these three values of $D_L^{eff}(t)$, it would be difficult to infer that the dispersion coefficient depends on the value of R/d.

Table 3 provides a comparison of the experimental data with the theoretical predictions, showing agreement in trend of D_L^{eff} with R/d. The experimental values are also plotted and compared with the predicted values from the extended Aris model in Figure 4a. The model underestimates experimental results for D_L^{eff} for the two smaller cylinders, R/d = 9.5 and 12.7, but the spacing between the two predictions is consistent with the spacing between the two experimental values. The model slightly overestimates D_L^{eff} for the largest cylinder, R/d = 25.4. However, the scaling of the predictions and the experimental values are consistent with a strong dependence on R/d and with the prediction that R^2/D_T is the relevant time scale for fully developed dispersion in cylindrical packed beds.

Conclusions

We have presented experimental evidence that for a given particle Peclet number, the longitudinal dispersion coefficient in a packed column of spheres depends on the column length and the ratio of column radius to sphere diameter, R/d. Dispersion coefficients were obtained in three columns of different radius using the same mean pore velocity and sphere diameter. The column lengths were determined from a theoretical requirement that $L > \bar{v}R^2/D_T$. The results show that the effective longitudinal dispersion coefficient depends on R/d, and that the dependence is consistent with the predicted time scale. The results are also consistent with the scaling of D_L^{eff} predicted by an extended Aris model of dispersion in a packed column.

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