Flow and Dispersive Behaviors of Cylindrical Pellet String Reactor

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INTRODUCTION

The measurement of rate parameters in heterogenous catalysis by gas chromatography has been recognized as a rapid and dependable means and had been reviewed by Smith et al. (1976). Scott et al. (1974) adopted a combination of gas chromatography and a rather special reactor which is packed only with single string of spherical pellets. The spherical pellet string reactor (SPSR) is convinced to have the same flow behaviors as conventional packed columns by two reasons. First, the pressure drop across SPSR follows the Ergun equation (1952) for conventional packed beds. Second, the SPSR has the same axial dispersive behavior as that of conventional beds. Chou and Hegedus (1980) use the SPSR coupled with pulse technique to measure diffusivity. However, these works are limited to spherically-shaped pellet and the widely-adopted, cylindrically-shaped pellet are not concerned. It is demonstrated by this study that a cylindrical pellet string reactor (CPSR) has behaviors similar to SPSR, if suitable packing pattern, flow rate, and column to pellet diameter are chosen.

EXPERIMENTAL

Pressure drop across the CSPR are read from a two-liquid manometer filled with water and benzylalcohol (or carbon tetrachloride). Commercial catalyst supports γ-Al_2O_3 with average 0.325 cm in diameter and 0.347 cm in length are packed in glass column of average inside diameter 0.495 cm. The axial directions of pellets are perpendicular to each other alternatively and all perpendicular to the axial direction of the column. The length of the column used is not less than 1 m, the longest one used for low Reynolds number is about 6 m in length.

To study the axial dispersion, the CPSR is connected to a six ports sampling valve and a thermoconductivity detector (TCD). All of them are thermostated. A short α-Al_2O_3-packed column is inserted between the CPSR and the TCD to delay the total residence time and match the response time constant of the recorder. The broadening of the response curves caused by nonimpulse tracer, dead space in the system, and the short packed column are removed by Fourier analysis and deconvolution technique adopted in the computer program for the dispersion coefficient search. The carrier gas is Matheson H.P. grade helium and a mixture of 2–3.5% nitrogen in helium is tracer. Nonporous brass pellets are used as packings.

RESULTS AND DISCUSSION

The main differences between CPSR and conventional packed bed are the packing pattern and the diameter ratio of pellet to bed. Since there is only single string of pellets in a CPSR, the wall effect must not be neglected. Scott et al. (1974) suggested that if the wall area is included appropriately in the definition of the hydraulic diameter in formulating dimensionless groups, the correlations applied to conventional bed can also adequately describe the behavior of SPSR. An equivalent diameter (d_m) can be defined according to the specific surface area in a CPSR as:

$$S_m = \frac{\text{total surface area of pellets and wall}}{\text{total volume of pellets}} = \frac{6}{d_m} \tag{1}$$

The Ergun equation is

$$\frac{\Delta P d_m e^3}{L_t \rho u^2 (1-e)} = K_1 \frac{\mu(1-e)}{u d_m \rho} + k_2 \tag{2}$$

For conventional bed $k_1 = 150$ and $k_2 = 1.75^{(3)}$. Data for CPSR are illustrated in Figure 1. As may be seen in the figure, k_1 is well predicted by the correlation. The k_2 value for SPSR obtained by Scott et al. (1974) was lower than 1.75 and a further lower value is found for CPSR in this study. The results seem reasonable. It is noted that the first term on the righthand equation is due to skin friction and the second term to kinetic energy loss. Since all the area presented to fluid are included in d_m, the first term fits well into its physical significance and k_1 is precisely predicted. But for the second term, the kinetic energy loss is caused by turbulent eddies of sudden enlargements and contractions in the cross-sectional area of the flow channels. The changes in cross-sectional area are mainly demonstrated by particles and particles in conventional bed which is large, and by particles and wall in CPSR or SPSR which is considerably smaller; thus, the decrease in k_2 is expectable. When CPSR is compared to SPSR, one may note that the enlargement and contraction caused by identical spherical particles and wall is larger than that by cylindrical pellets and wall. This is the reason for further decrease in k_2 in CPSR. Although deviation in high Reynolds number region exists, if the Fourier analysis experiment is carried out at rather low flow rate, say $N_{Re} < 100$, in which the skin friction dominates, the basic argument that the CPSR has the same flow behaviors as conventional packed bed is still well proved.

With the exception of adopting deconvolution technique to eliminate the effect other than CPSR, the same procedures rec-

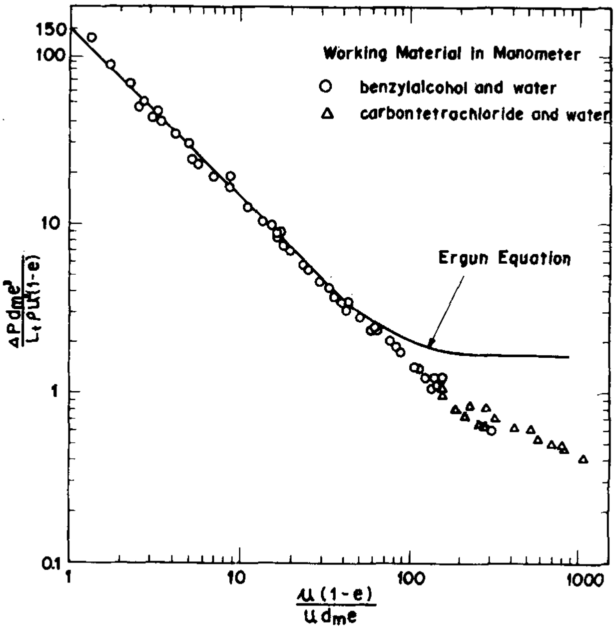

Figure 1. Pressure drop of CPSR

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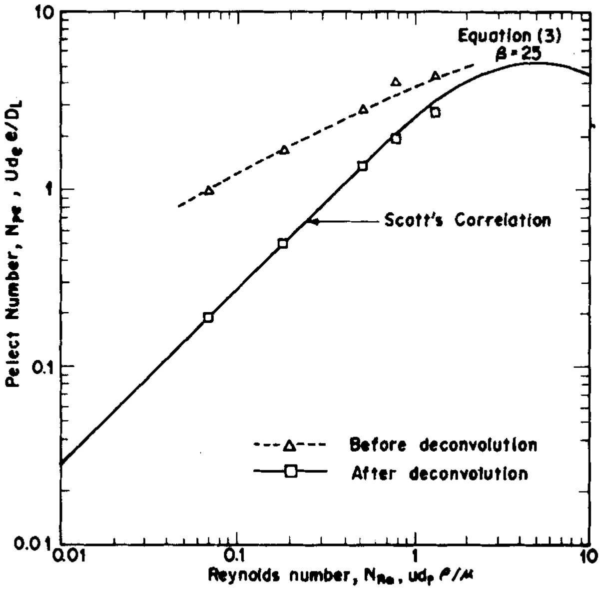

Figure 2. Test of Confidence level with SPSR

ommended by Scott are used to search for the axial dispersion coefficients of the CPSR. To check the effectiveness of the deconvolution technique and the confidence level of our system, SPSR packed with steel ball is also studied before the CPSR experiments. The results are excellent in comparison with Scott's data. These are shown in Figure 2.

Edward and Richardson (1968) correlated the axial dispersion coefficients by equation.

$$N_{Pe}{}^{-1} = \frac{\gamma}{N_{Re}N_{Sc}} + \frac{\lambda}{1 + \dfrac{\beta}{N_{Re}N_{Sc}}} \tag{3}$$

γ, λ and β values from different sources and this work are listed in Table 1 for comparison. d_e is defined as six times the ratio of pellet volume to surface. The brass pellet used in this work has average value $Lp = 0.348$ cm, $dp = 0.330$ cm, and $de = 0.332$ cm. Two column diameters (diameter ratio of column to pellet = 1.54, 1.81) and two types of packing pattern, one with the axial directions of pellets all perpendicular to the column axis, and the other with the axial directions of pellets perpendicular and parallel alternatively to the column axis, are tested. The axial dispersion coefficient is correlated well by Eq. 3, Figure 3. The value is related to the tortuosity of packed bed; higher value means a lower tortuosity. It is just the case for CPSR as shown in Table 1.

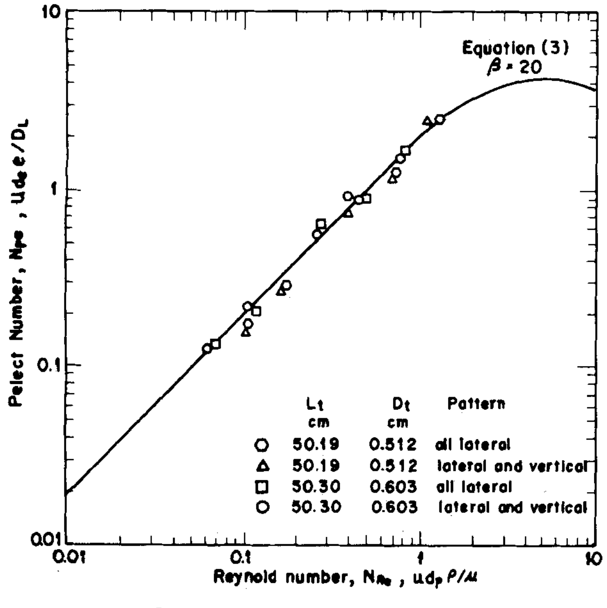

Figure 3. Axial dispersion in CPSR

TABLE 1. COMPARISON OF PARAMETERS IN EQ. 3

	System Tracer-Carrier	γ	λ	β	N_{Sc}
Edwards & Richardson	Air-Argon	0.73	0.5	9.7	0.72
Urban & Gomezplata	He-N_2	0.73	0.5	14.1	0.35
Scott, Wey Lee & PAPA	H_2-N_2	0.75	0.5	22.0	0.205
(Spherical Pellet Above)	N_2-He	0.64	0.5	25.0	1.662
	He-N_2	0.57	0.5	24.1	0.222
This Work	N_2-He	0.86	0.5	20.0	1.650

CONCLUSION

The experimental results of this work suggest that, with suitable design, CPSR can be used to find the physical parameters within and around a cylindrical pellets through pulse technique and Fourier analysis.

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NOTATION

D_{AB} = binary diffusivity
D_L = true axial dispersion coefficient
d_e = equivalent diameter of cylindrical pellet, 6 volume/surface area
d_m = equivalent diameter or characteristic diameter of CPSR
dp = diameter of cylindrical pellet
d_t = diameter of glass tube column
e = void fraction of CPSR
k_1,k_2 = parameters in Ergun equation
Lp = length of cylindrical pellet
L_t = length of CPSR
N_{pe} = Pelect number, $udee/D_L$
N_{Re} = Reynolds number, $ude\rho/\mu$
N_{Sc} = Schmidt number, $\mu/\rho D_{AB}$
ΔP = pressure drop across CPSR
S_m = specific surface area of CPSR
u = average interstitial velocity
β,γ,λ = parameters in Eq. 3
ρ = density of carrier gas

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A General Empirical Relationship between Tracer or Self-Diffusion Coefficients of Liquids and Pressure

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In the course of recent work on tracer diffusion of HTO and carbon-14 labelled CH_3CN, in $H_2O + CH_3CN$ mixtures under pressure, it was found to be useful to represent the variation of diffusion coefficients with pressure (at constant temperature) in such a way as to facilitate comparison of the diffusion behavior of the mixtures. Testing a variety of analytical relationships between D and P has led to the discovery of a simple relationship which linearises D,P data over wide pressure ranges, and which holds at widely differing temperatures. The relationship has been tested for a considerable number of tracer and self-diffusion coefficients as a function of pressure and in all but a few cases (noted below), and except for pressures close to normal atmospheric pressure, it represents experimental data to within the probable limits of uncertainty of the data.

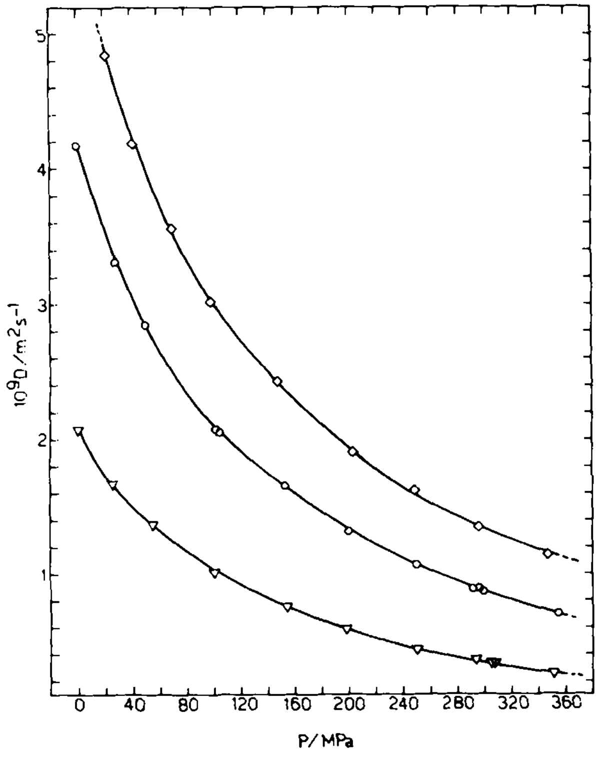

Figure 1. The variation with pressure of the self-diffusion coefficient for n-hexane. ◇, 333.15 K; ○, 298.15 K; ▽, 248.15 K.

The relationship has the simple form

$$\ln D = a + b\,P^{0.75} \qquad (1)$$

and is potentially useful in engineering situations because it allows prediction, to within the uncertainty of the experimental data, of diffusion coefficients at high pressures from as few as two values at lower pressures. A limitation of this relationship is that in many cases it overestimates values of D at pressures near 0.1 MPa. However, since diffusion coefficients are more readily available at 0.1 MPa then at high pressures, the applicability of the relationship at high pressure is an advantage which outweighs the disadvantage of inapplicability at low pressure. Furthermore, the discrepancy between experimental and predicted diffusion coefficients at or near normal atmospheric pressure decreases with increasing temperature.

The recently determined self-diffusion coefficient data for n-hexane (Harris, 1982) provide a stringent test for Eq. 1 because the data have relatively high precision (±1.5%), and they are available up to high pressures (maximum 393.8 MPa) and cover a wide temperature range (from 223.15 to 333.15 K). The data for three

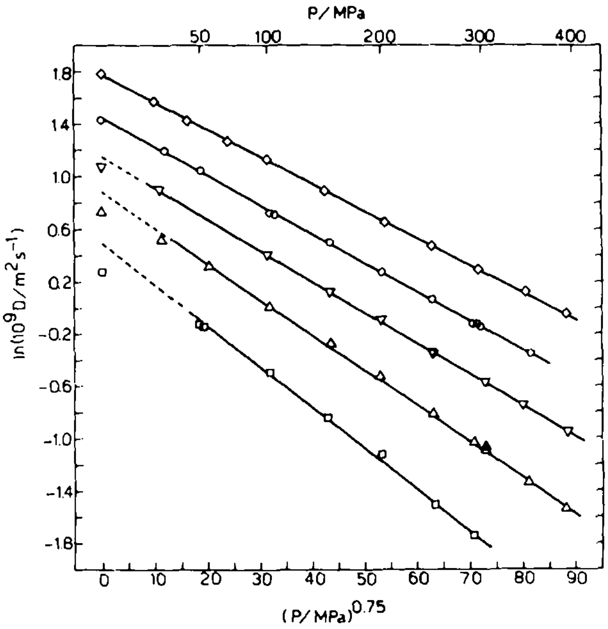

Figure 2. Fit of Eq. 1 to self-diffusion data for n-hexane. ◇, 333.15 K; ○, 298.15 K; ▽, 273.15 K; △, 248.15 K; □, 223.15 K.