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...... UNION CARBIDE CORPORATION NUCLEAR DIVISION



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SUBJECT:

Dispersion of Miscible Fluids in Porous Media, Part 2

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ABSTRACT

Miscible-liquid displacements of water/ethanol solutions were conducted in a 2.54-cm-ID column packed with glass beads of 275-300 and 25-30 mesh with bed heights of 6.35 to 40.8 cm. The viscosity ratio of the resident fluid to displacing fluid was maintained at 0.72 for fluid pairs with favorable density differences. Dispersion coefficients obtained from a two-parameter model increased from $\sim\!\!3$ x 10^{-5} to $\sim\!\!2$ x 10^{-2} cm²/sec with increasing packing particle size, fluid velocity, and density difference.

> Oak Ridge Station School of Chemical Engineering Practice Massachusetts Institute of Technology S.M. Senkan, Director

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SUMMARY

The phenomenon of miscible-liquid displacement in porous media exists in tertiary oil recovery and liquid chromatography. When the fluid to be displaced is more viscous than the displacing fluid, the more mobile, displacing fluid may protrude into the resident phase in "viscous fingers." Similarly, in vertical flow, "gravity tongues" may develop when the top fluid is denser than the bottom fluid. Such conditions arising from density and viscosity differences are termed "unfavorable," indicating a potential for greatly increased dispersion.

Miscible-liquid displacements of water/ethanol solutions were conducted in a 2.54-cm-ID column packed with glass beads of 275-300 and 25-30 mesh. Bed heights were varied from 6.35 to 40.8 cm to determine the importance of end effects and to develop a correlation for dispersion coefficients in doubly favorable displacements. The ratio of displacing-fluid-to-resident-fluid viscosities was maintained near 1.35 for three different fluid pairs with 0.0125, 0.0125, and 0.022 qm/cc density differences.

End effects were present only in displacements of the largest density difference in beds of small particles. Dispersion coefficients, obtained from a two-parameter model, increased with increasing favorable density difference, velocity, and particle diameter. However, absolute density or viscosity had no apparent effect. Dispersion coefficients were correlated with both the Reynolds number of the displacing fluid and the difference between Reynolds numbers. Future studies should investigate different ranges of densities and viscosities to ascertain the correct functional form of this correlation.

2. INTRODUCTION

2.1 Background

Miscible-liquid displacement in porous media is of importance because of its applications in tertiary oil recovery, chromatography, and leaching of buried radioactive wastes. Densities and viscosities of the phases influence dispersion in this process. If the resident fluid is more viscous than the displacing fluid, then the more mobile, less viscous displacing fluid may protrude into the resident phase, causing viscous fingering. Similarly, gravity tongues may develop in density-unfavorable vertical displacements in which the top fluid is denser than the bottom fluid.

Several models have been proposed to simulate this process (7): the dispersion model predicts symmetric S-shaped concentration profiles, and the capacitance model (3) accounts for "stagnant" volumes in the porous media, giving asymmetric concentration profiles with tailing. Mixing cell, film, and statistical models have also been suggested (7), but no satisfactory model has been proposed to simulate displacements with the highly irregular

concentration profiles caused by extremely unfavorable viscosity or gravitational conditions.

Previous workers (4) have attempted to correlate dispersion coefficients with critical velocity. Which is the velocity at which pressure gradients were equal or the velocity at which the interface is allegedly stable. Using Darcy's Law, this velocity is given by

$$v_{c} = \frac{\beta_{r} - \beta_{d}}{\mu_{r} - \mu_{d}} \tag{1}$$

In vertical flow, gravity forces dominate at low velocities, so viscosity unfavorabilities may exist without causing fingering. A low critical velocity is then predicted by Eq. (1). At high velocities, inertial and viscous forces jominate, and even a small unfavorability in viscosity may cause fingering. Equation (1) then predicts an upper value for V_c . When fluid pairs have identical viscosities and densities, Eq. (1) loses its meaning. In two previous MIT projects, dispersion in favorable and unfavorable displacements using a variety of packing materials and liquid pairs was investigated (2, 6). It was found that dispersion coefficients increased with velocity and particle size for displacements with symmetrical concentration profiles

2.2 Objectives

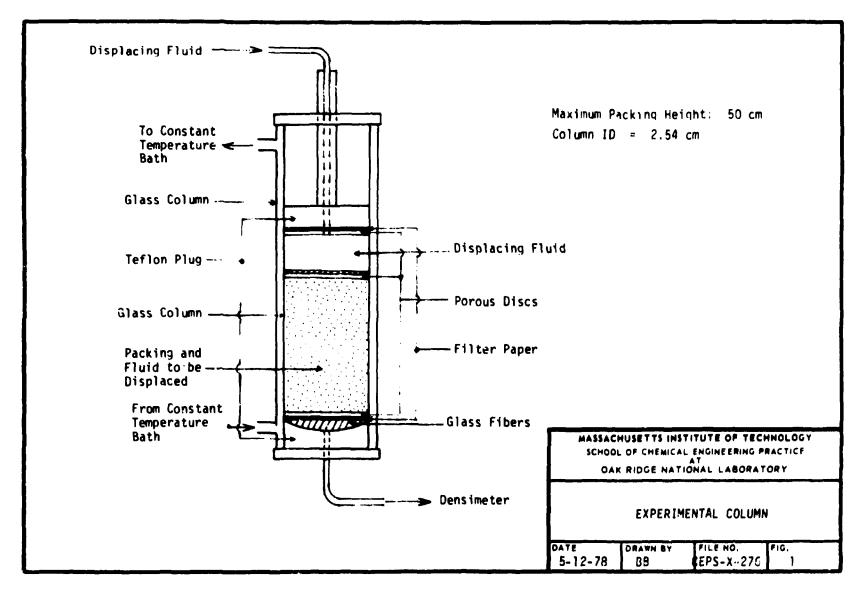
The objectives were: (1) to determine whether entrance and exit effects are significant in a 2.54-cm-ID column, and if so, to eliminate them by modifying the apparatus, and (2) to correlate dispersion coefficients in doubly favorable displacements as a function of the control variables, such as liquid densities and viscosities, particle diameters, and flow rates.

3. APPARATUS AND PROCEDURE

3.1 Experimental

Experiments were conducted in a 2.54-cm-ID x 50-cm-long column (Fig. 1), packed with spherical glass beads of either 0.0049 cm (275-300 mesh) or 0.065 cm (25-30 mesh) in diameter. To minimize entrance effects, the displacing fluid was introduced through a small Teflon tube into the space between a plunger and a porous-disk flow-distributor just above the packing. A rubber 0-ring was installed above the disk to minimize wall channeling. A concave plug with glass fiber packing at the bottom of the column was also used to minimize exit effects. Temperature was kept at 13°C by a circulating





water bath. Pore volumes were calculated conventionally from the weight and volume of glass beads packed in the column.

Four liquid pairs, all mixtures of ethanol and water, were used (Table 1). Liquid flow rates from 1.2 to 24 ml/min were used, with bed heights ranging from 6.35 to 40.8 cm. The density of the effluent stream was monitored by a SODEV 02D densimeter.

table 1. Experimental rivid Pairs							
Fluid Pair	% EtOH	ρ (gm/cc)	<u>µ (mp)</u>	∆o (gm/cc)	μ_{d}/μ_{r}		
displacing	7.25	0.9850	12.95	-0.0125	1.32		
A resident	0	0.9975	9.80	-0.0125			
displacing B	16.25	0.9725	17.50	0.0125	1.35		
resident	7.5	0.9850	12.95	-0.0125			
displacing	31.25	0.9500	24.15	0.0225	1.42		
C resident	16.25	0.9725	17.00	-0.0225			
_displacing D*	0	0.9975	9.80	.0.0475	0.41		
resident	31.25	0.9500	24.15	+0.0475			

Table 1. Experimental Fluid Pairs

3.2 Mathematical Model

The effluent density or concentration profile was matched to the dispersion model. Assuming no radial gradients, the model simulates dispersion as:

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

If an infinite core is assumed, the boundary and initial conditions are set as follows:

at
$$x = 0$$
, $vC_0 = vC - D \frac{\partial C}{\partial x}$ (3)

^{*} Unfavorable both in density and viscosity.

as
$$x + \infty$$
 $C(x,t) + 0$ (4)

at
$$t = 0.0$$
 $C(x,0) = 0$ (5)

The solution to Eq. (2) using Eqs. (3), (4), and (5) is then

$$\frac{C}{C_0} = \frac{1}{2} \operatorname{erfd}(\frac{\sqrt{\gamma}}{2} \frac{y-I}{\sqrt{I}}) - \frac{\sqrt{I}}{\sqrt{\pi \gamma} (y+I)} e^{-\gamma (y-I)^2/4I} (1-2\frac{I}{1+I})$$
 (6)

where:

 $\gamma = vL/D$

y = dimensionless length, x/L

I = pore volume injected or the dimensionless time, vt/L

If one considers the existence of relatively stagnant packets in the bed, e.g., molecular diffusion dominates, the solution becomes:

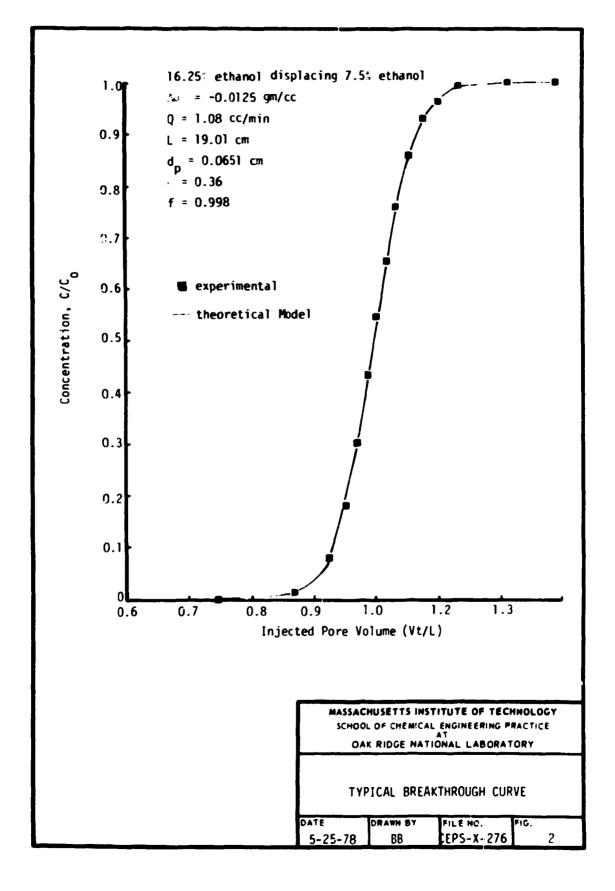
$$\frac{C}{C_0} = \frac{1}{2} \operatorname{erfc}(\frac{\sqrt{\gamma}}{2} \frac{y-J}{\sqrt{J}}) - \frac{\sqrt{J}}{\sqrt{\pi \gamma}(y+J)} \exp[-\gamma(y-J)^2/4J] (1 - 2 \frac{J}{1+J})$$
 (7)

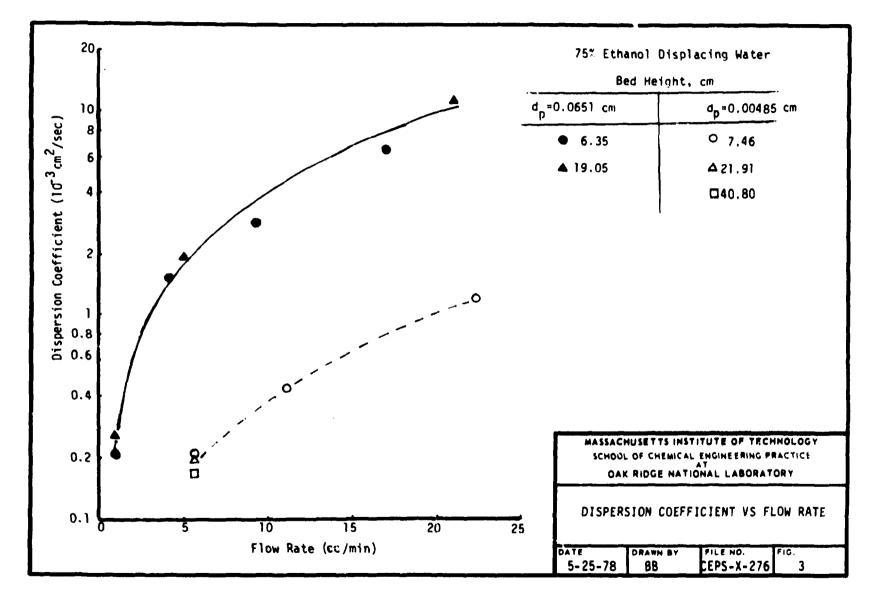
where J = I/f and f is the fraction of the bed which is active. Chow and Alger (1) have developed a computer model based on Eq. (7), which was also used in this study.

4. RESULTS AND DISCUSSION

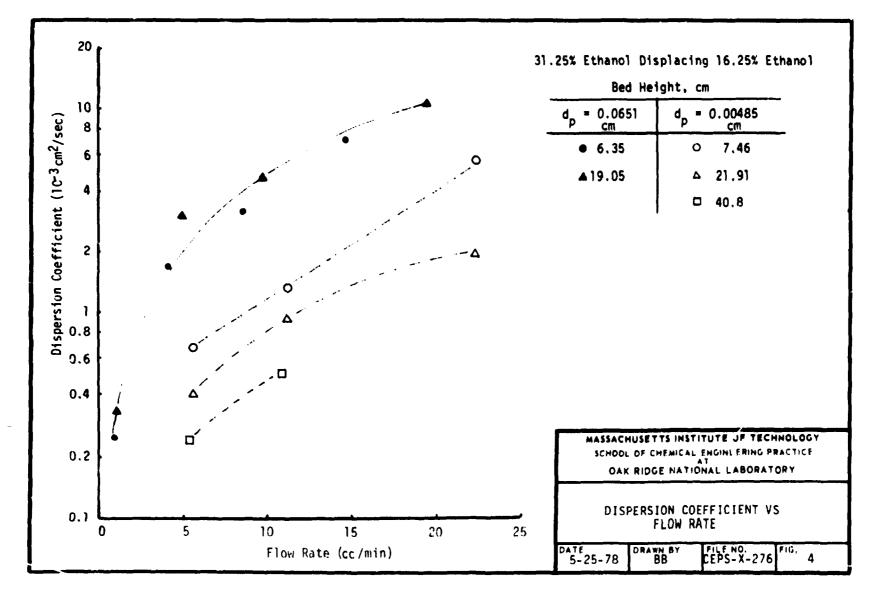
Figure 2 shows a typical symmetrical S-shaped concentration profile obtained from the effluent of a stable displacement and the best dispersion-model fit. In general, the dispersion model described the data well for symmetrical displacements. A summary of experimental displacements and model fits is included in Appendix 8.1.

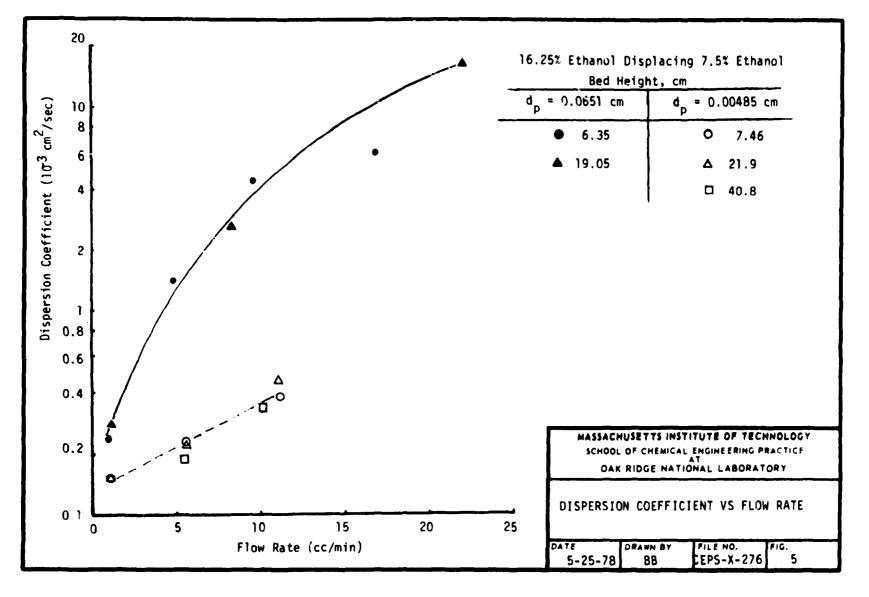
Figures 3, 4, and 5 show that end effects are negligible except in the displacement of the largest density difference in small packing (Fig. 4) in which the dispersion coefficient apparently increased with decreasing bed height. Figures 3, 4, and 5 show an increase in dispersion coefficient with increasing particle size, flow rate, and density difference. This is in agreement with previous workers, who also found the dispersion coefficient to increase with particle size and flow rate (2, 6). As velocity increases the convective contribution to dispersion increases, and therefore a greater dispersion coefficient is obtained.











Krupp's data (5) agree with these findings in that dispersion increases with increasing favorable density difference. However, this is contrary to what was reported by Howlett (4), that mixing length decreases as density difference becomes more favorable. If dispersion is gravity-controlled, gravitational forces should act to segregate the phases, decreasing the dispersion coefficient. However, if the flow regime is such that inertial viscous forces dominate, higher dispersion coefficients result from increases in density difference. Dispersion coefficients were about equal for displacements with fluids of equal density differences and viscosity ratios but different absolute values.

Krupp's results (5) and the results of this study can be unified if D is plotted as a function of Red, Reynolds' number of the displacing fluid (Fig. 6). Two distinct zones exist. For Red > 10^{-3} , dispersion is high enough that molecular diffusion is negligible. For Red < 10^{-3} , the dispersion coefficient is the same order of magnitude as diffusion coefficients; therefore the dependence of D on Red is weaker than in the first zone.

The correlation shown in Fig. 6 was found unsatisfactory as D was dependent only on the properties of the displacing fluid. D was also plotted as a function of $\mathrm{Re_r}$ - $\mathrm{Re_d}$, and a similar correlation was obtained (Fig. 7). The slopes of the lines in Figs. 6 and 7 are nearly the same, suggesting that other, perhaps more meaningful, combinations of $\mathrm{Re_d}$ and $\mathrm{Re_r}$ can be used to correlate the data.

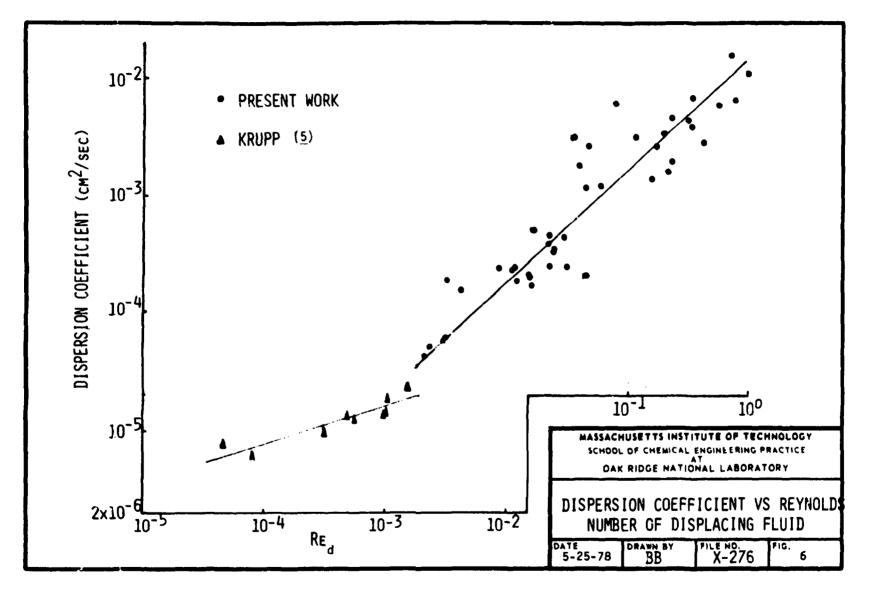
5. CONCLUSIONS

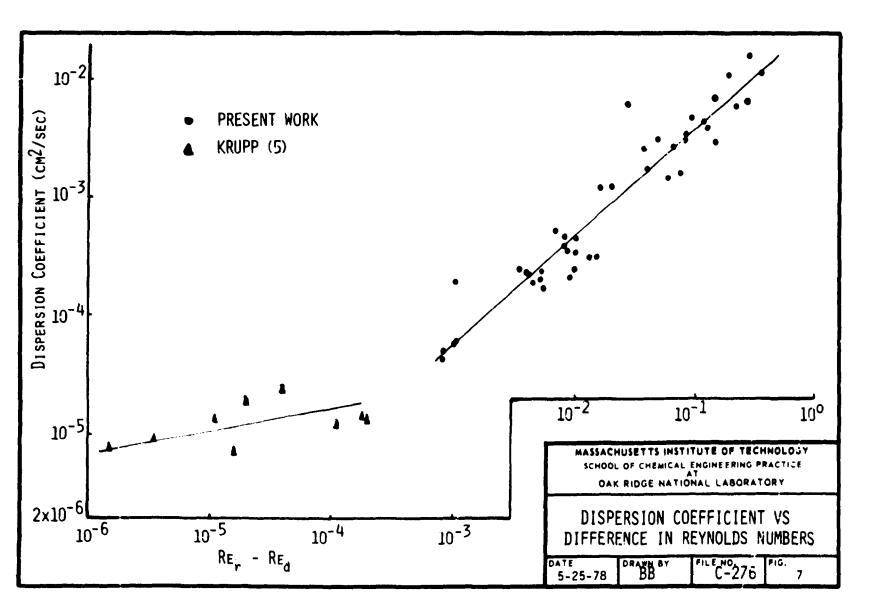
- 1. The dispersion coefficient was proportional to Re_d and $(\mathrm{Re}_\mathrm{r}$ $\mathrm{Re}_\mathrm{d})$.
- 2. For $Re_d > 10^{-3}$, molecular diffusion was insignificant compared with dispersion.
- 3. End effects were enhanced by large density difference and small particle size.
- 4. Absolute densities and viscosities had no apparent effect on the dispersion coefficient.

6. RECOMMENDATIONS

It is clear that some form of Reynolds number is the important factor in correlating dispersion coefficients. Future studies should investigate different ranges of densities and viscosities to ascertain the correct functional form of this correlation.







7. ACKNOWLEDGMENT

The authors thank their consultants, S.Y. Shiao, K.A. Kraus, and J.S. Johnson, for their guidance and helpful suggestions throughout the project.

8. APPENDIX

8.1 Summary of Results

Table 2 presents the summary of experimental conditions and results. Following Table 2 are experimental breakthrough curves showing the fit of the theoretical model. The dimensionless concentration $(\mathsf{C}/\mathsf{C}_0)$ at the outlet is plotted as a function of injected pore volumes or dimensionless time.

8.2 Location of Original Data

The original data were recorded in ORNL Databook A-8144-G, pp. 46-62, and on chart paper in the calculation files of the MIT School of Chemical Engineering Practice, Bldg. 3001, ORNL.

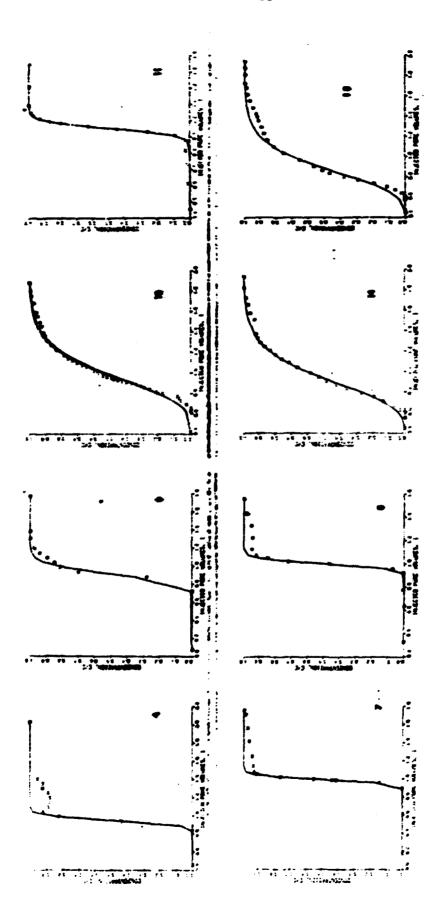
8.3 Nomenclature

- A cross section of column
- C concentration, presented only as a ratio, C/C_o
- D dispersion coefficient, cm²/sec
- $\mathbf{d}_{\mathbf{D}}$ particle diameter, cm
- f active bed fraction
- g gravitational constant
- K permeability (Darcy)
- L bed height, cm
- I pore volumes injected , Vt/L
- J I/f
- Q volumetric flow rate, cc/min
- Re Reynolds number, $\rho vd_{\mathbf{D}}/\mu$
- t time, sec
- v interstitial velocity, cm/sec, $Q/A\varepsilon(60)$

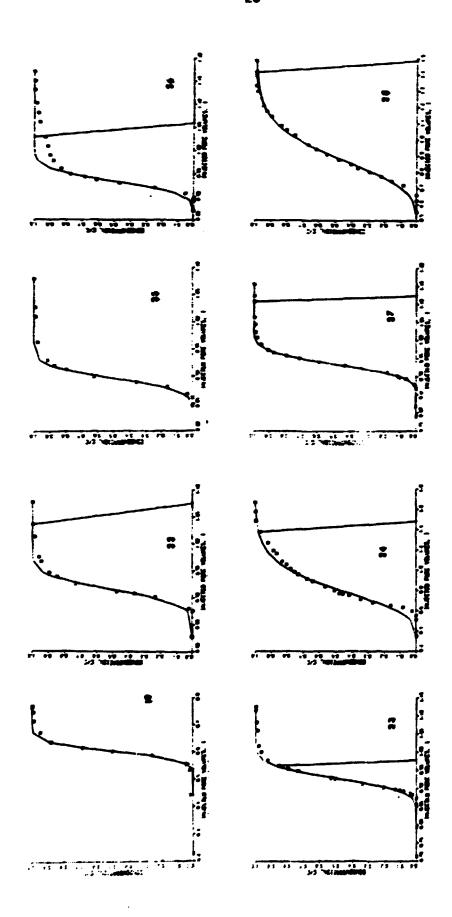
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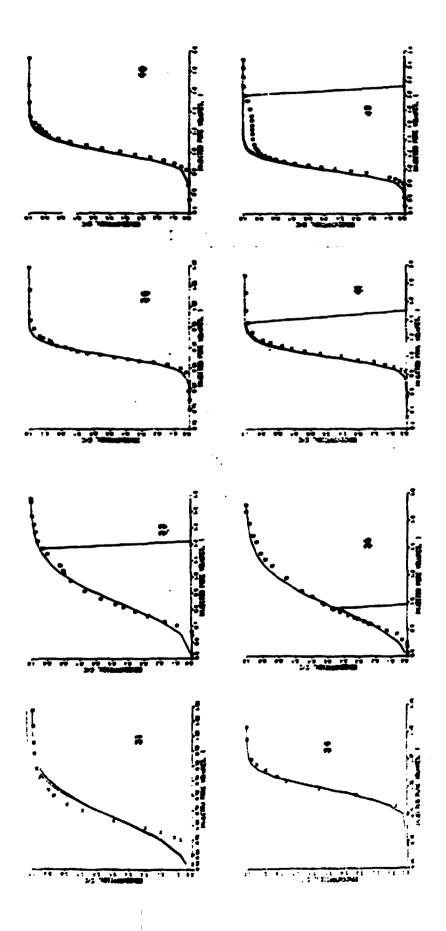
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-	Fluid Pair (Befor to					_			
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27 28	C D			4.019±10 ⁻⁴ 1.5620±10 ⁻²	9. 9300 9. 906 7	0.0 58 0 0.0206	8,0113 9, 900 2	-0.0033 -0.0126	
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62	Ċ	14.69	0.1353	6.9272±10 ⁻³	:	0.3665	0.4095	≈0,1430	
43 88	D A	14.63 9.31	0.1716 0.0858	2.3907±10 ⁻² 2.8420±10 ⁻³	1	1 , 137T 17, 4248	0,4 394 0, 566 5	-0. 0977 +0.1437	
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47	Ď	9.57	0.0002	7.8623-10-7	i	0.504	0.2759	-0.3505	
48 49	Á	4,72 4,81	3.9635 0.9666	1.5303e10-3 1.3099e10-3	1	0.2156 0.1696	0.2 0 92 0.2199	=0.0728 =0.0593	
50	ζ	4,11	0.0379	1.6916±10-1 3.4621±10-7	0.9496	2.0973	0,3371	rg. 9400	
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56 57	Å	21. 66 22.16	0.1790 0.2036	1.1037±10° ² 1.5731±10° ²	0.8100	9.7306	1.0082	+9.2716	
58	C	19.32	0.1776	1.9616x10*7	1	0.4548	1,6425	+9.1877	
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68	A	1.05	9.0096	2.5785±10-4	0.9958	0.0475	1.7636	+9.0161 +0.0122	
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74	č	10.73	0.7886	5.000mx1/1-4	0.1768	0.0169	1.0739	+0.0070	
16	A	5.72	0.0429	1.667±10-6	0.4999	0.0:58	9,4212	of ,0054	
7.7 78	8 C	5, 46 5, 46	0.0449 0.0449	1,9663x10+6 2,41:36x10+6	0.9566 0.9795	0.0121 2.0006	3.01 66 7.9121	+0,0965 +0,0935	
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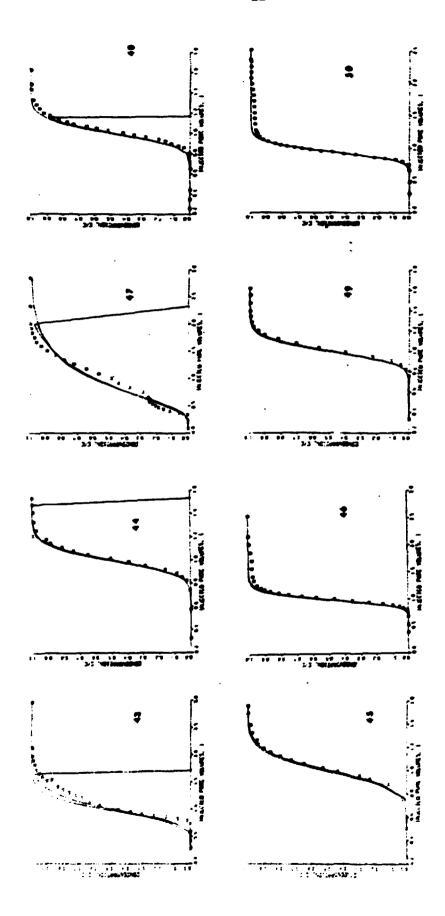
[&]quot;Where no f-value is given, D was calculated assuming f = 1.

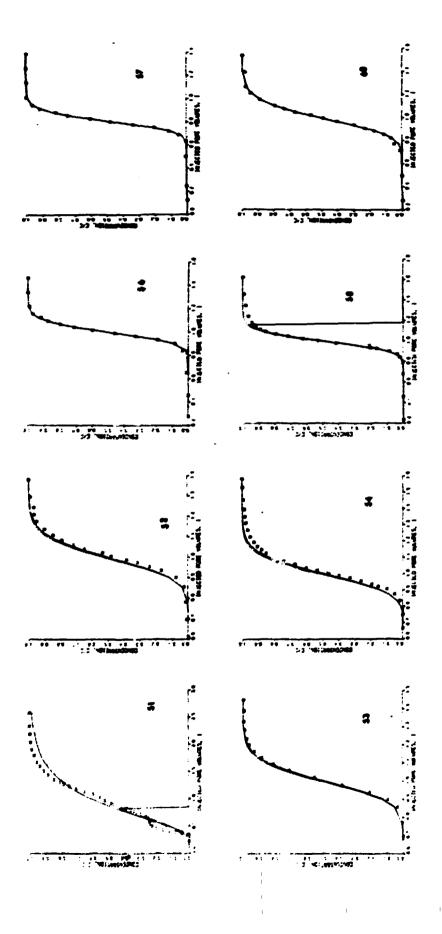


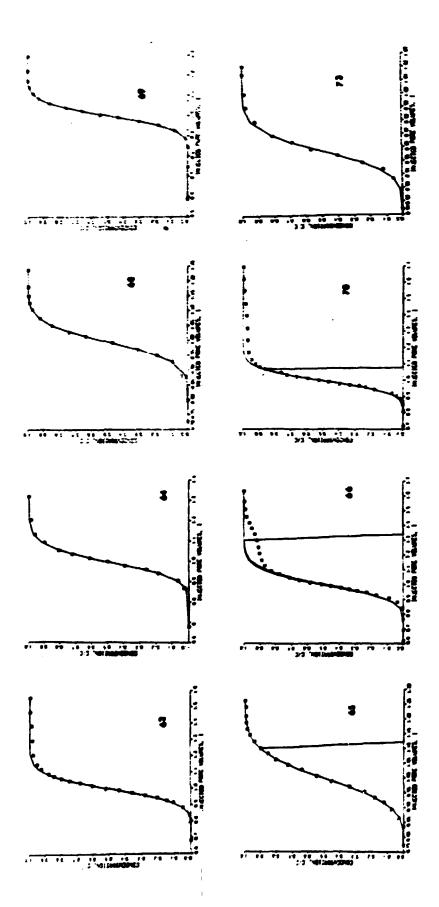
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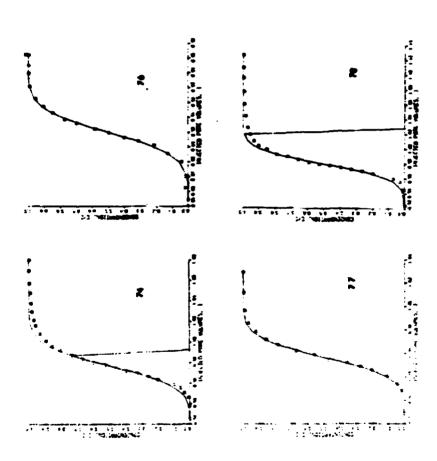












- v critical velocity, cm/sec
- x position in axial direction, cm
- y dimensionless length, x/L

Greek symbols

- Y VL/D
- ε void fraction in bed
- u viscosity, mp
- ρ density, gm/cc

Subscripts

- d displacing fluid
- o initial
- r resident fluid

8.4 Literature References

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