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ORNL/MIT-259

DATE: October 19, 1977

SUBJECT: Dispersion of Miscible Fluid in Porous Media

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ABSTRACT

The effect of particle diameter, density difference, and fluid flow rate on the stability of miscible displacements were investigated using ethanol and water solutions in a 50-cm x 2.54-cm-ID column packed with spherical glass beads. The flow characteristics were adequately described by a modified dispersion model that involves the dispersion coefficient, D, and the fraction of the bed that was active, f.

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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy $4.50 ; Microfiche $3.00

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1. SUMMARY

Miscible displacement in a packed bed of spherical glass beads was studied using water and ethanol solutions of different concentrations. Density differences, particle diameters, and flow rates were varied to isolate the onset of unstable displacement characterized by widening of the mixing zone between displacing and displaced fluids.

When a denser fluid displaces a lighter one in downflow, the mixing length may be broadened by formation of protrusions of the denser fluid into the less dense fluid at low velocities. The displacement will be unstable when gravity effects disrupt the symmetrical concentration profiles in the mixing zone between the two fluids. Instabilities became more pronounced with increased packing sizes, increased density differences, and decreased flow rates. The transition from stable to unstable displacement as indicated by the mixing length was gradual.

A dispersion model which uses the dispersion coefficient, D, and the fraction of mobile fluid, f, as parameters (2) described the column behavior adequately.

2. INTRODUCTION

2.1 Background

Dispersion accompanying the flow of miscible fluids through porous media occurs in oil recovery, chromatographic techniques, and leaching of buried radioactive wastes. Previous researchers (2, 3, 5, 7, 8) have developed theoretical models describing the dynamics of miscible displacement in porous media. Based on Darcy's Law, empirical correlations and perturbation methods have been utilized to determine system parameters which adequately characterize flow stabilities/instabilities and dispersion. Most studies have examined instabilities due to viscous effects, with very few studies evaluating the effects of density differences.

According to Darcy's Law, the velocity in a porous medium is given by:

$$U = -\frac{K}{\mu}\left[\left(\frac{dP}{dz}\right) - \rho g\right] \tag{1}$$

for a single phase system. Dumore (4) and Slobod and Howlett (9) have shown that for stable miscible displacement P and dP/dz will be equal at the interface. Therefore, using Eq. (1) the critical velocity is

$$U_c = \left[\frac{\rho_1 - \rho_2}{\mu_1 - \mu_2}\right]Kg \qquad (2)$$

This criterion correlates experimental observations accurately; however, it fails to yield meaningful results if either the density or viscosity of the liquids was kept constant.

For a downflow miscible displacement, four distinct cases may exist: density favorable-viscosity favorable, density unfavorable-viscosity favorable, density favorable-viscosity unfavorable, and density unfavorable-viscosity unfavorable. When a highly dense liquid displaces a lower density liquid, protrusions known as gravity tongues may form, thus broadening the interface (see Fig. 1). The opposite case may have the stabilizing effect on the mixing zone. Viscosity fingers may form when displacing liquid is less viscous than resident liquid, again broadening the mixing zone (see Fig. 1). The converse is true for a higher viscosity displacing liquid. Higher liquid velocities tend to suppress gravity effects; however, they may increase the viscosity induced instabilities.

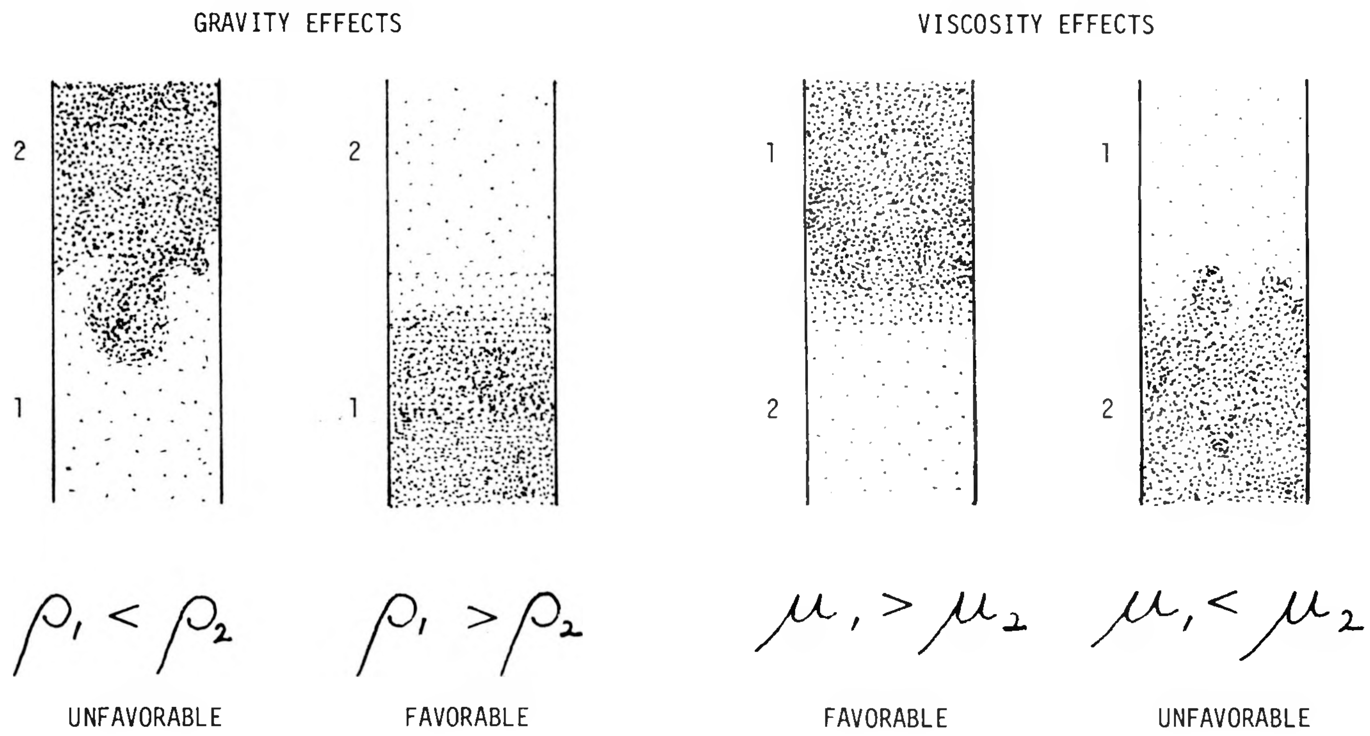
A recent MIT group (1) evaluated miscible dispersions using various packing materials and concluded that dispersion increased with increasing liquid velocities and particle sizes at fixed liquid densities and viscosities.

2.2 Objectives

The objectives were to investigate the effects of particle size, liquid flow rate, and density difference on the nature of miscible displacements (6) and to evaluate the applicability of a two-parameter dispersion model for stable displacements (2).

3. APPARATUS AND PROCEDURE

Liquids used for displacement experiments were mixtures of ethanol and water. This system was chosen because mixtures of equal viscosities, yet different densities, were easily obtained (Fig. 2). Table 1 shows the pairs of fluids used in the experiments. Two pairs of fluids close to the maximum viscosity composition were chosen to minimize the viscosity peak at the mixing zone. One pair was chosen with a large density difference to check for effects of this phenomenon by comparing results of density favorable and unfavorable displacements.



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FAVORABLE AND UNFAVORABLE DISPLACEMENT

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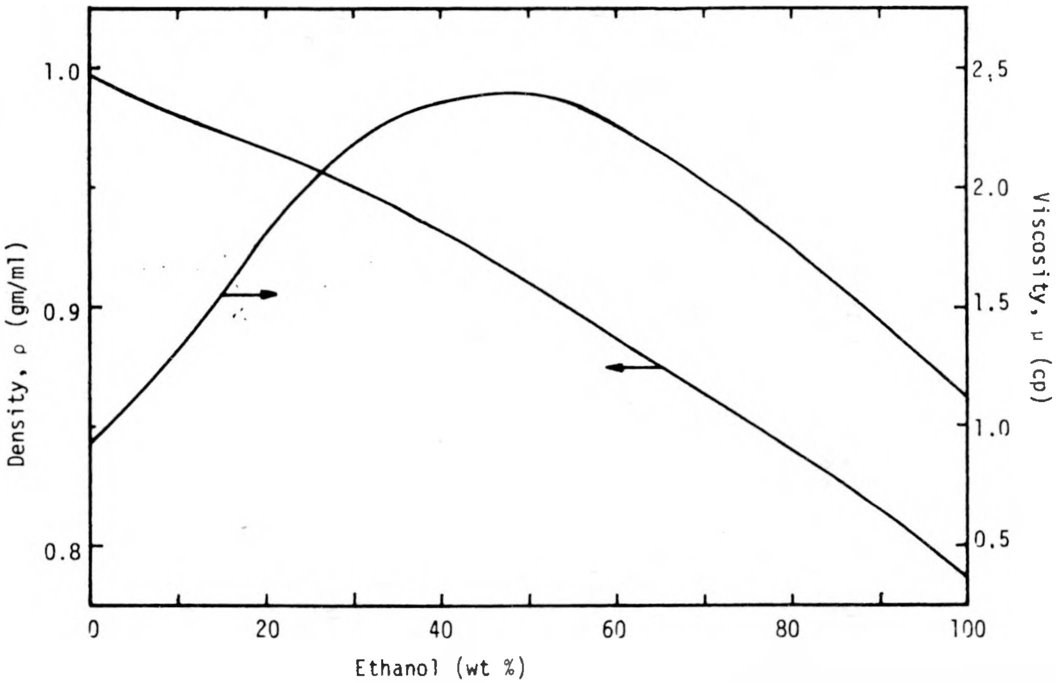

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DENSITY AND VISCOSITY OF ETHANOL/WATER MIXTURES (1)

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10-18-77	RSC	CEPS-X-252	2

Table 1. Experimental Fluid Pairs

wt % EtOH	ρ (gm/ml)	μ (cp)
24.5	0.960	2.0
71.0	0.862	2.0
36.25	0.940	2.3
60.0	0.895	2.3
38.75	0.934	2.35
54.0	0.903	2.35

Experiments were conducted in a vertical column (Fig. 3), 2.54-cm-ID x 50-cm, packed with spherical glass beads of size ranges: 0.125-0.149, 0.250-0.297, and 0.595-0.707 mm. The column was first charged with one liquid, and then the flow was switched to the displacing fluid at the top of the column. Flow rates ranged from 1 to 10 ml/min. Column and feed solutions were kept at 25°C by a circulating water bath. The density of the effluent was monitored by a SODEV 02D densimeter (Fig. 4). Pressure drop and pore volume through the column were also measured. Pore volume was determined by using the mean residence times and by weighing the column and its contents.

4. RESULTS AND DISCUSSION

Plots of dimensionless outlet concentration vs time were used to distinguish between stable and unstable cases. Unstable cases show multiple inflections indicating an irregular concentration profile, whereas stable curves show smooth S-curves (see Fig. 5).

4.1 Breakthrough Curves

A summary of experimental results are presented in Table 2. The runs with favorable density differences produced stable displacements at the two lower density differences. For the highest density difference, the density-favorable runs showed instability for the small and medium packing sizes which can be attributed to viscosity effects. Consequently results at the greatest density difference are of little value in determining the effects of density alone on stability.

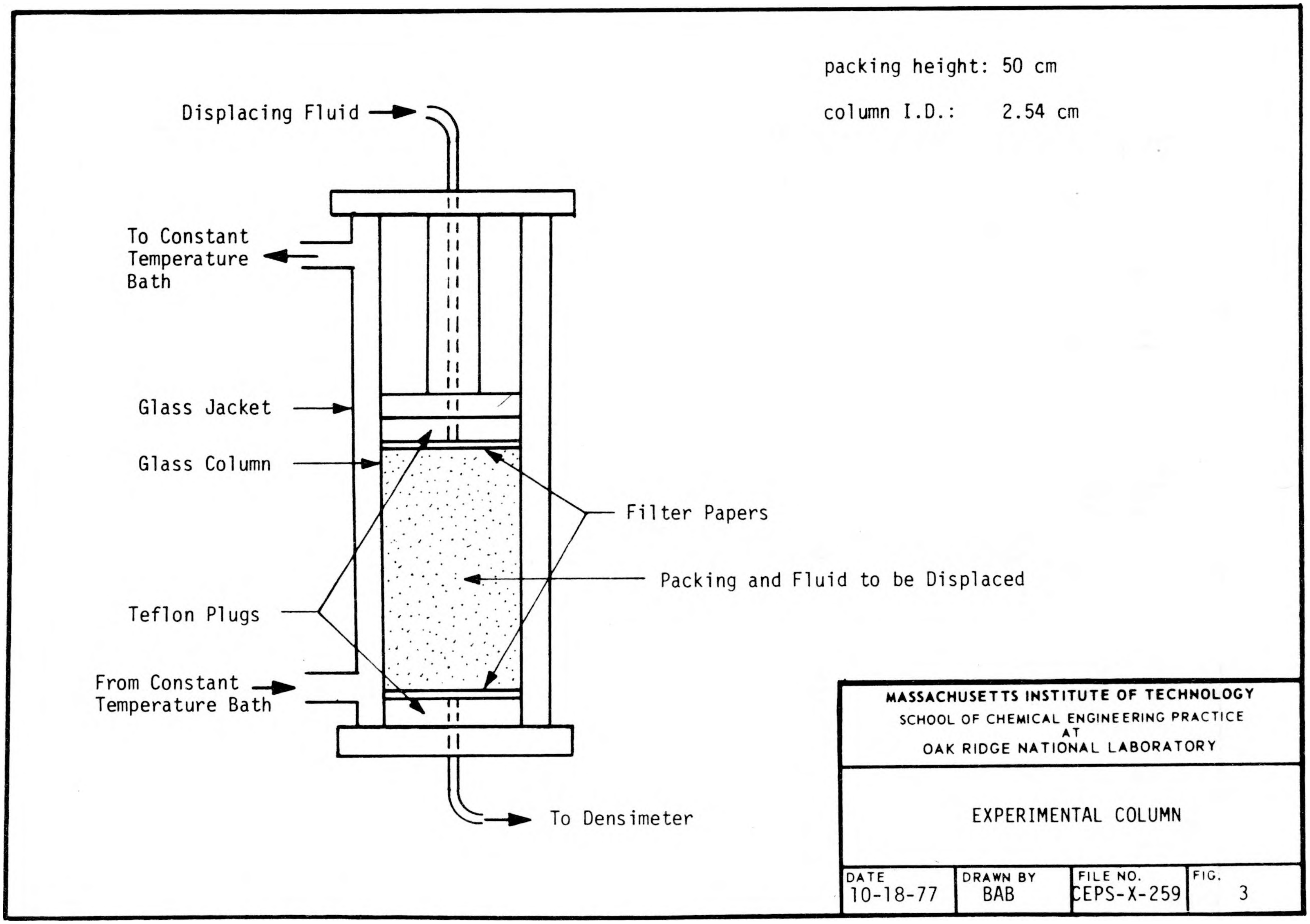
packing height: 50 cm
column I.D.: 2.54 cm
Displacing Fluid
To Constant Temperature Bath
Glass Jacket
Glass Column
Filter Papers
Packing and Fluid to be Displaced
Teflon Plugs
From Constant Temperature Bath
To Densimeter
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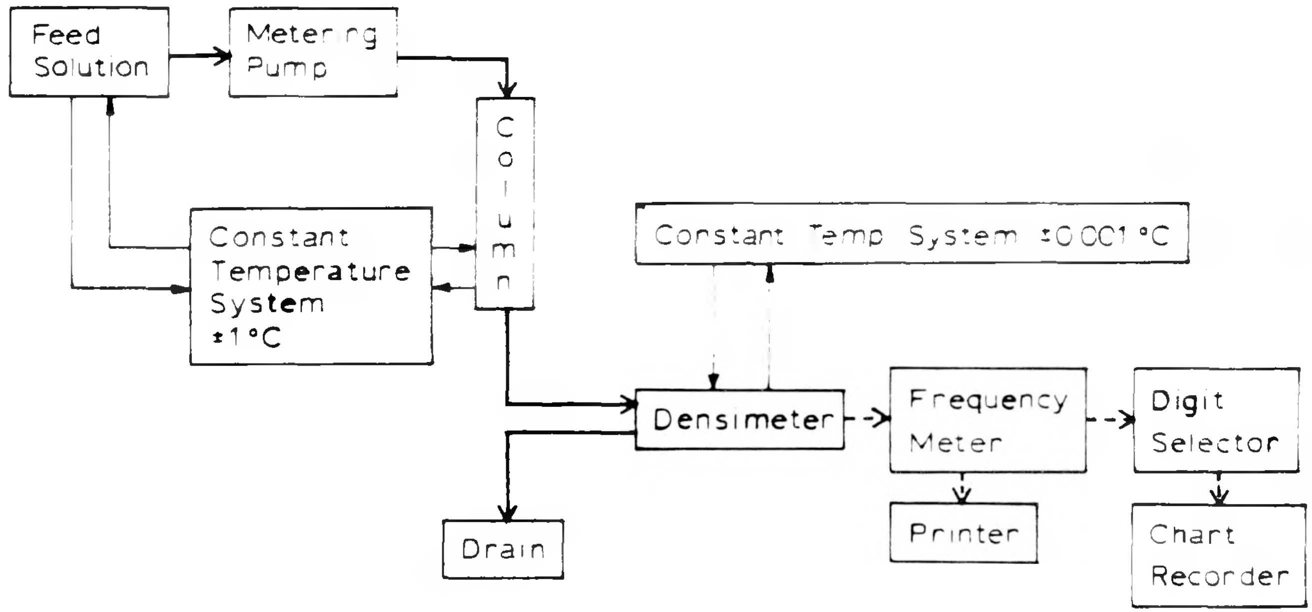
FIG. 3



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SCHEMATIC OF EXPERIMENTAL APPARATUS (1)

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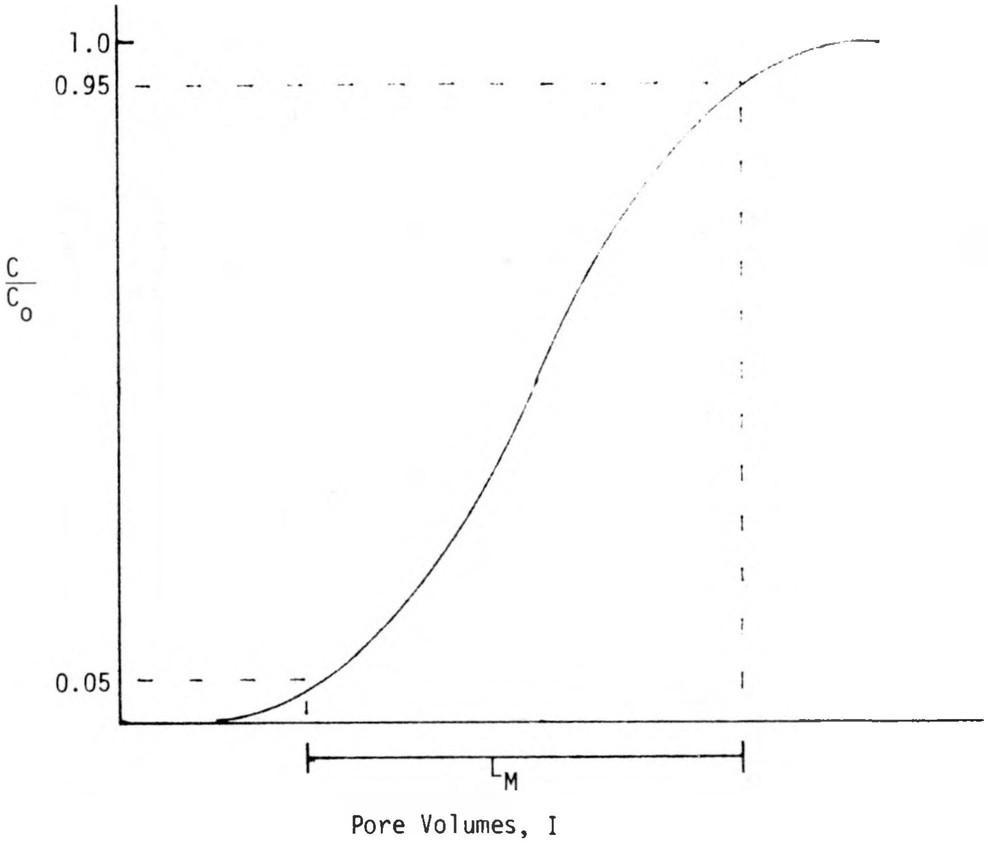

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BREAKTHROUGH CURVE INDICATING THE MIXING LENGTH DEFINITION

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Table 2. Observed Stability of Breakthrough Curves

Particle Size of Packing	Flow Rate (ml/min)	Density Difference (U), $\Delta\rho$ (gm/ml)[b] 0.031	0.045	0.098
d_p = 0.0610 cm	2			UNSTABLE
25-30 mesh	5		(UNSTABLE)[1]	
	10	STABLE		
d_p = 0.0262 cm	1			
50-60 mesh	2	(STABLE)		
	5	STABLE		UNSTABLE
	10			
d_p = 0.0130 cm	1			
100-120 mesh	2	STABLE		
	5			UNSTABLE
	10			

[1]Parentheses indicate that observation was inconclusive.

[b]Viscosity effects were evident for high density difference displacements for small and medium packing.

The runs with unfavorable density differences produced both stable and unstable displacements. For the two large packing sizes, a transitionary behavior was observed. For the largest packing size, the transition flow rate was between 2 and 5 ml/min for the smaller density difference and between 5 and 10 ml/min for the medium density difference. This trend was due to gravity effects becoming more noticeable as flow rates decreased and/or density differences increased (see Table 2).

For the medium packing size, transition from stable to unstable displacements was poorly characterized. The 1-2 ml/min runs made at the smallest density difference appeared to be unstable but well-behaved. At medium density difference, transition again occurred at higher flow rates.

The transition flow rate increased with increasing density difference and particle size. In large packings, interstitial spacing was wide enough to permit formation of gravity tongues even at high velocities. For small packings, gravity tongues did not form even at the slowest flow rates investigated.

4.2 Mixing Length

The definition of mixing length, L_M, is based on the cumulative exit age distribution function (see Fig. 5). Plots of mixing length vs flow rate give good indications of the stability/instability of miscible displacements. Figures 6 and 7 show the effect of liquid flow rates and density differences on mixing lengths at a fixed packing size. Similarly, Figs. 8 and 9 show the effects of flow rates and packing sizes on mixing lengths for a constant density difference. For stable cases the mixing length increases with increasing velocity due to increased micromixing. Conversely, for unstable cases, the mixing length decreases with increasing velocity since increased velocity reduces the time available for gravity tongues to develop; thus the protrusions of displacing liquid into the displaced liquid are minimized. For displacements which could not be classified as stable or unstable, mixing lengths were unaffected by flow rates.

4.3 Dispersion Coefficient

Matching experimental concentration profiles with those predicted by the dispersion model (2) permit calculation of an effective dispersion coefficient, D, and the fraction of unstagnant fluid, f, for both stable and/or unstable flow regimes. A least squares computer algorithm was successfully used in calculating these parameters. In all runs f was very close to unity.

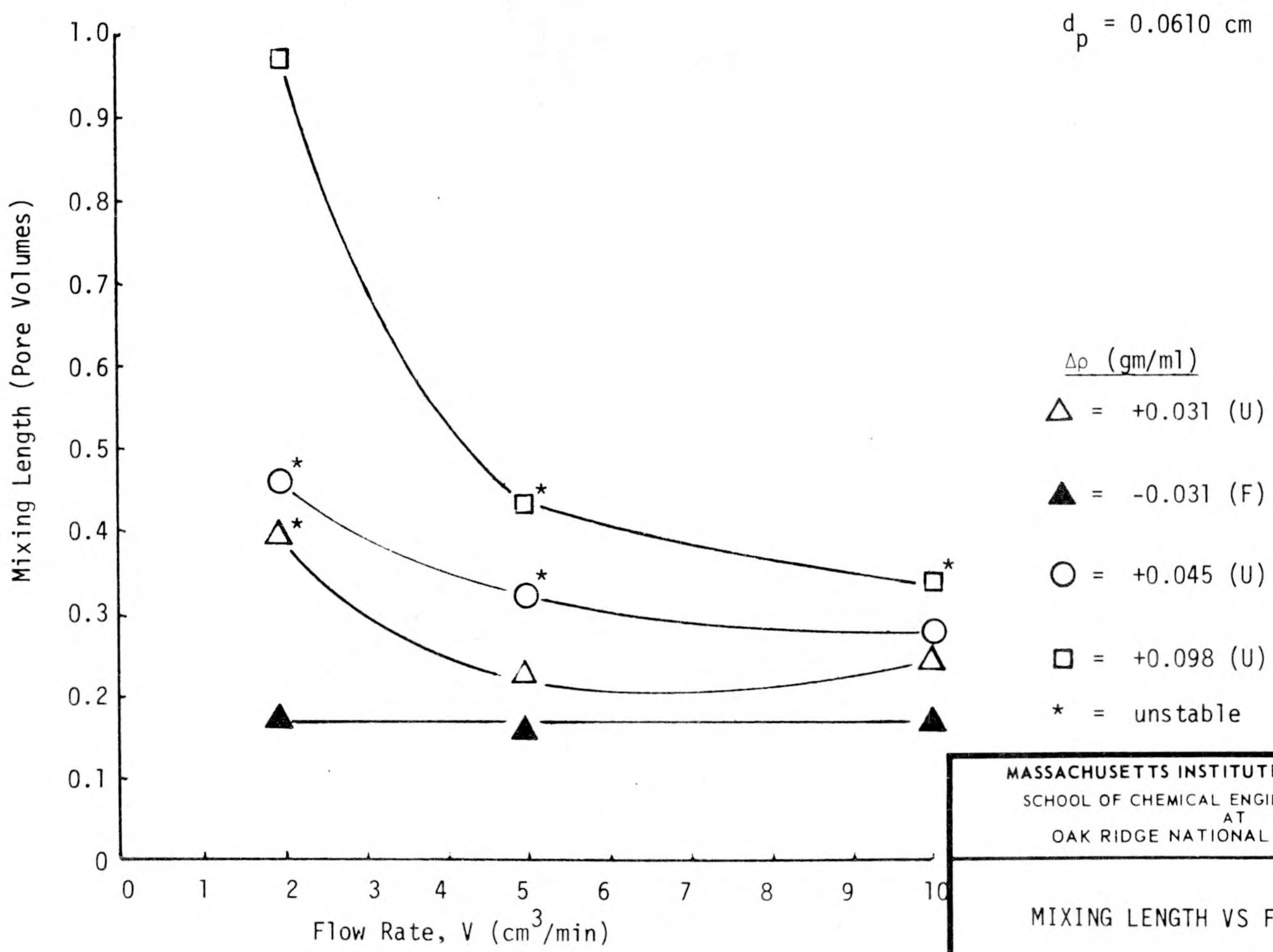
For stable flows, dispersion coefficient increased almost linearly with interstitial velocities (see Figs. 10, 11, and 12) in accordance with the trends predicted by the Taylor dispersion model (2). However, for unstable cases the dispersion coefficient actually decreased with increasing velocity due to the shorter residence times. Above a critical velocity, U_c, where stable flows occur, dispersion coefficients again increased linearly with velocity. Increasing density differences increased the dispersion coefficient for stable but unfavorable cases. However, dD/dU remained constant indicating a possible linear relationship between Taylor dispersion coefficient and the observed dispersion coefficient, D.

5. CONCLUSIONS

1. The transition from stability to instability, as measured by the spread of the cumulative exit age distribution function, appeared gradual.

2. For unstable cases, mixing length decreased with increasing flow rates; for stable cases, the converse was true.

3. The dispersion coefficient increased with increasing flow rates for stable displacements, while it decreased for unstable cases.



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MIXING LENGTH VS FLOW RATE			
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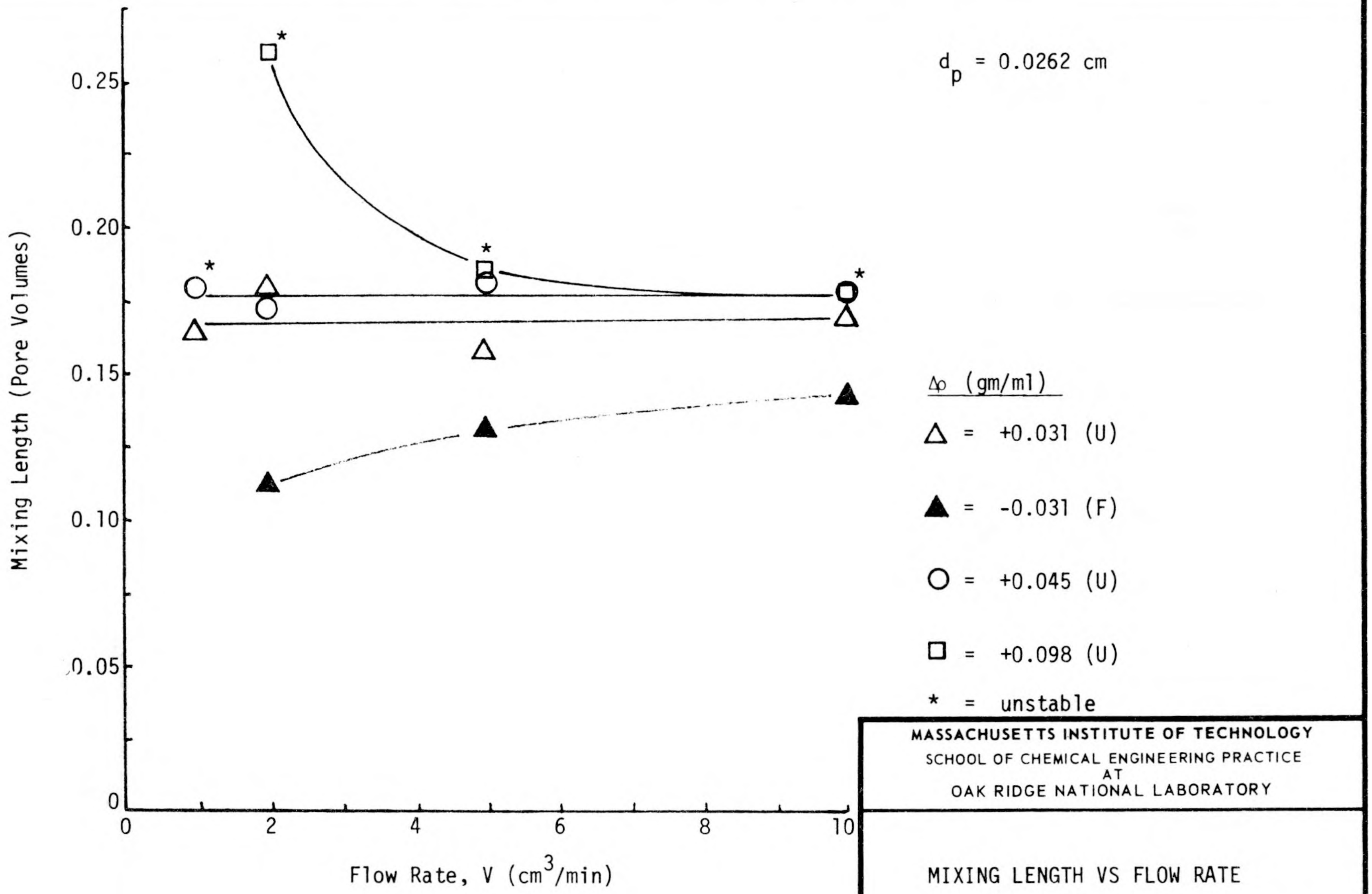

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MIXING LENGTH VS FLOW RATE

DATE	DRAWN BY	FILE NO.	FIG.
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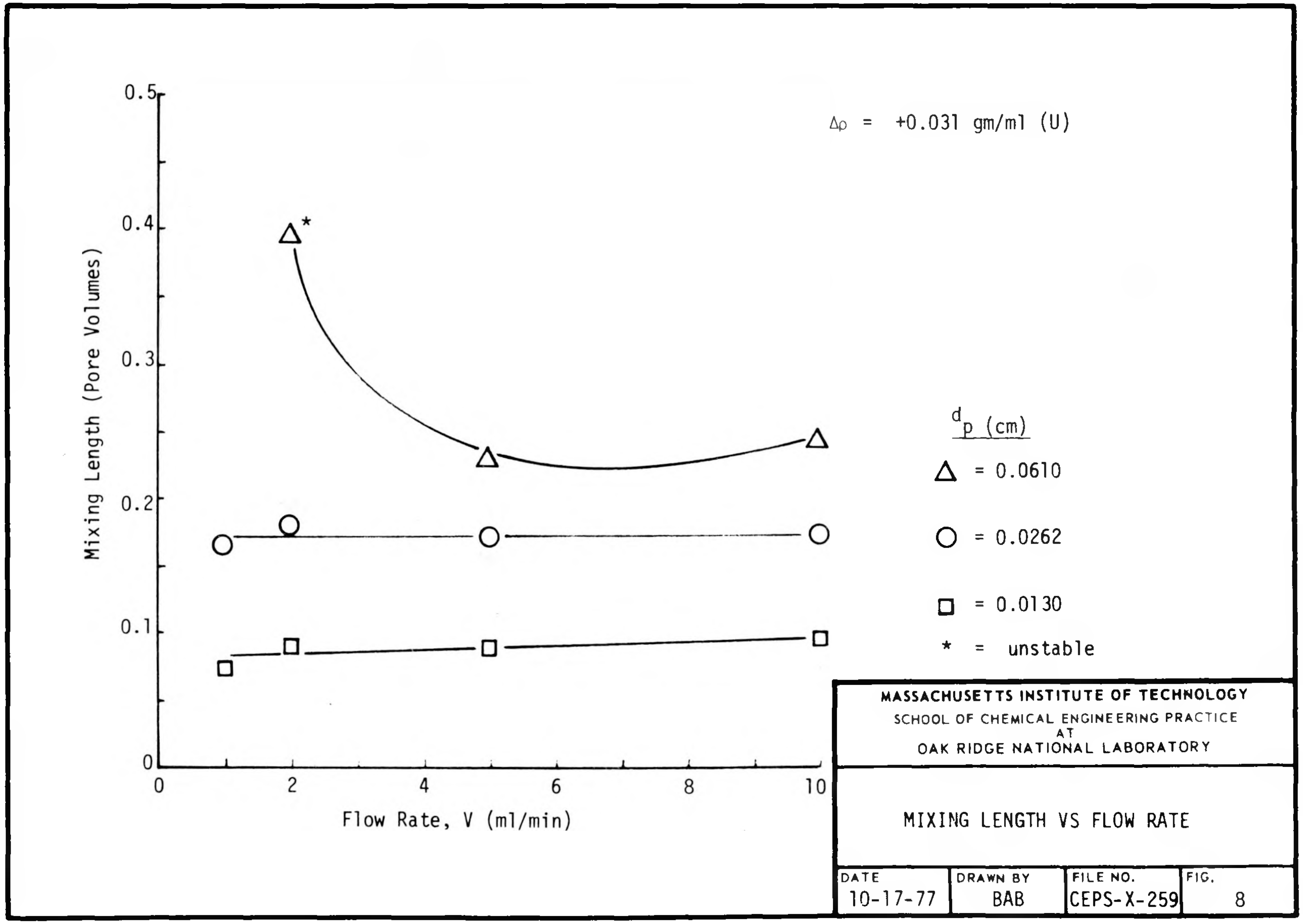

Mixing Length (Pore Volumes)
0.5
0.4
0.3
0.2
0.1
0
0
2
4
6
8
10
Flow Rate, V (ml/min)
Δρ = +0.031 gm/ml (U)
d_p (cm)
△ = 0.0610
○ = 0.0262
□ = 0.0130
* = unstable
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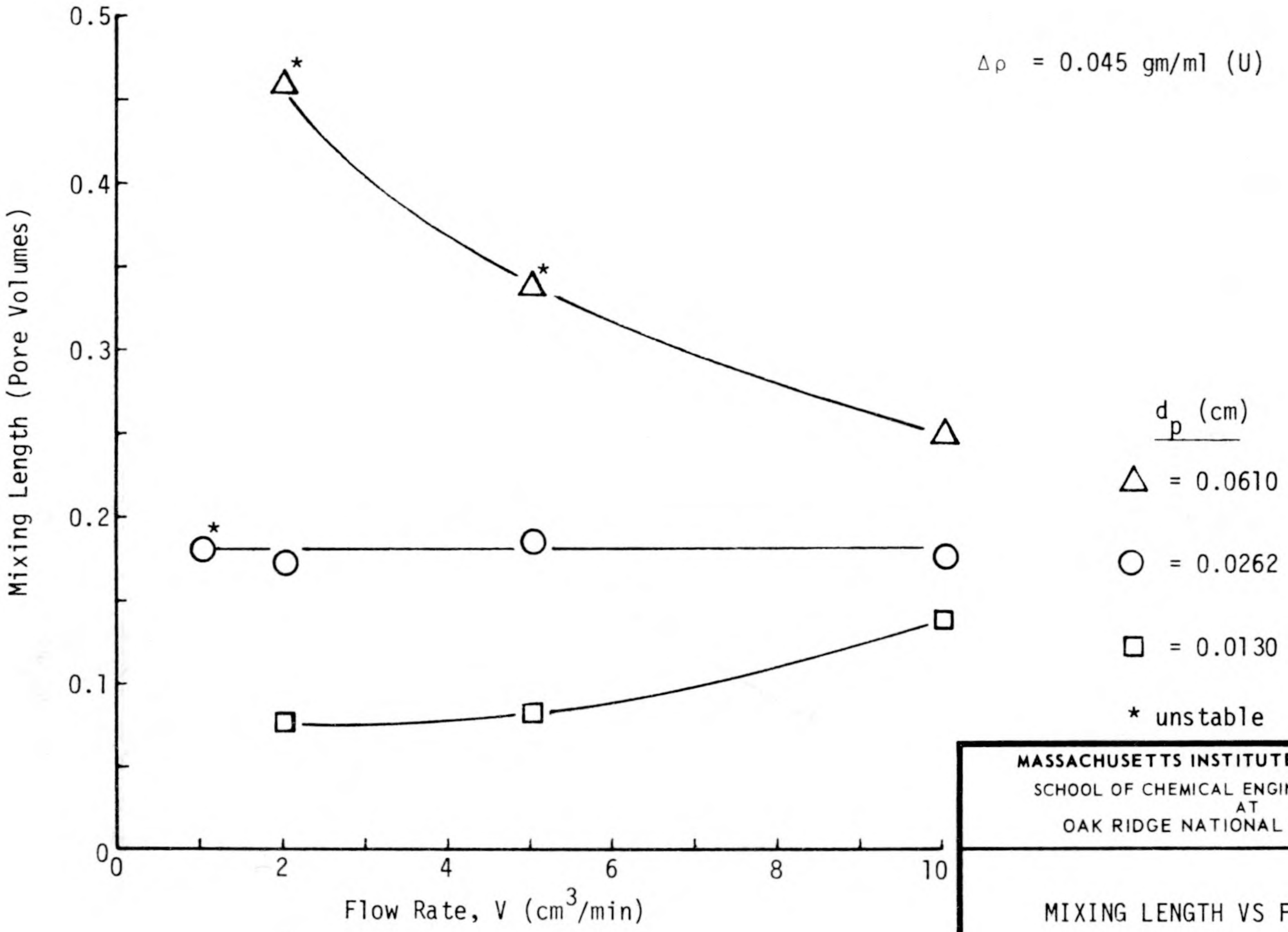
FIG. 8



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MIXING LENGTH VS FLOW RATE

DATE	DRAWN BY	FILE NO.	FIG.
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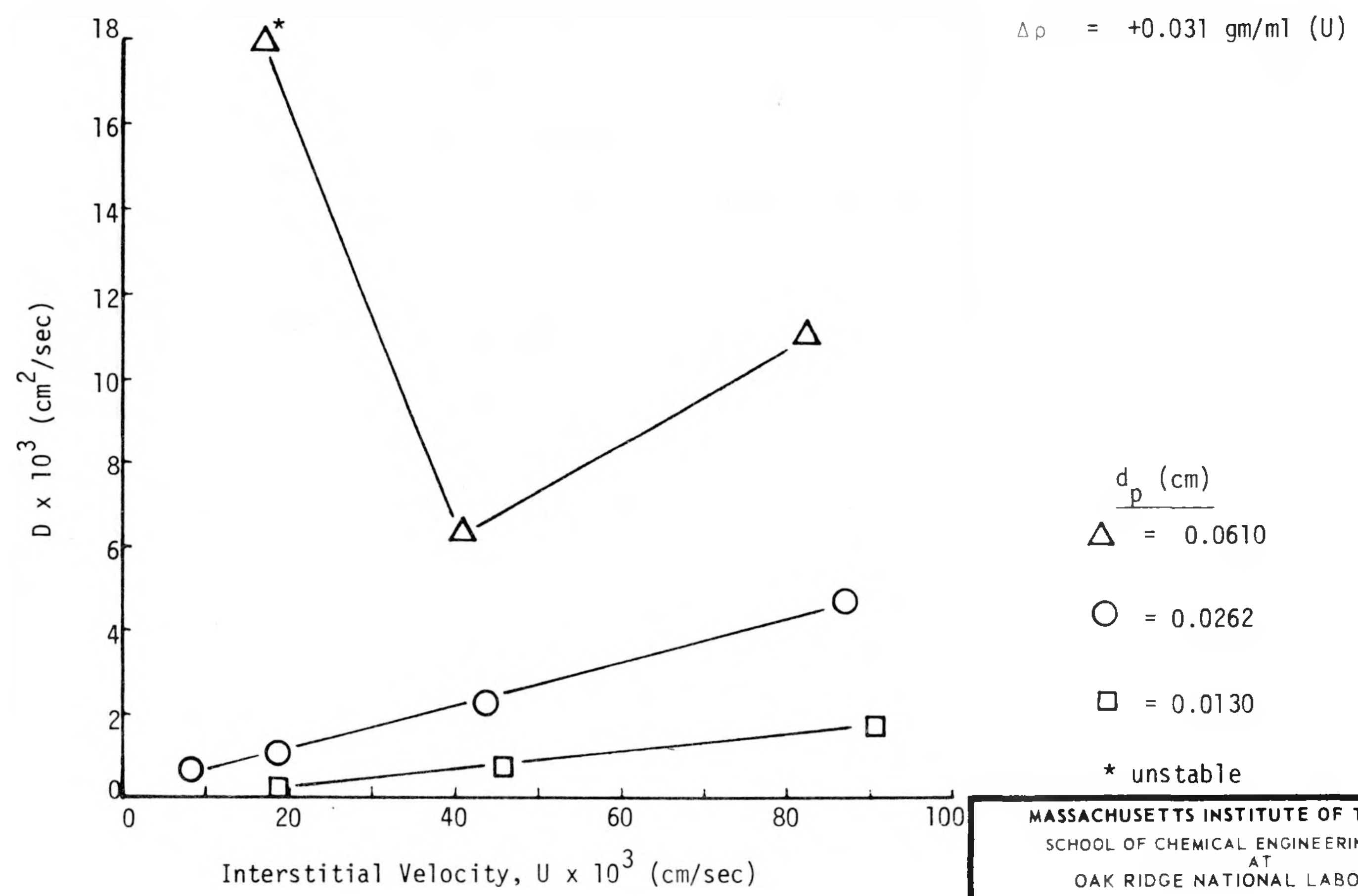

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DISPERSION COEFFICIENT
VS INTERSTITIAL VELOCITY

DATE	DRAWN BY	FILE NO.	FIG.
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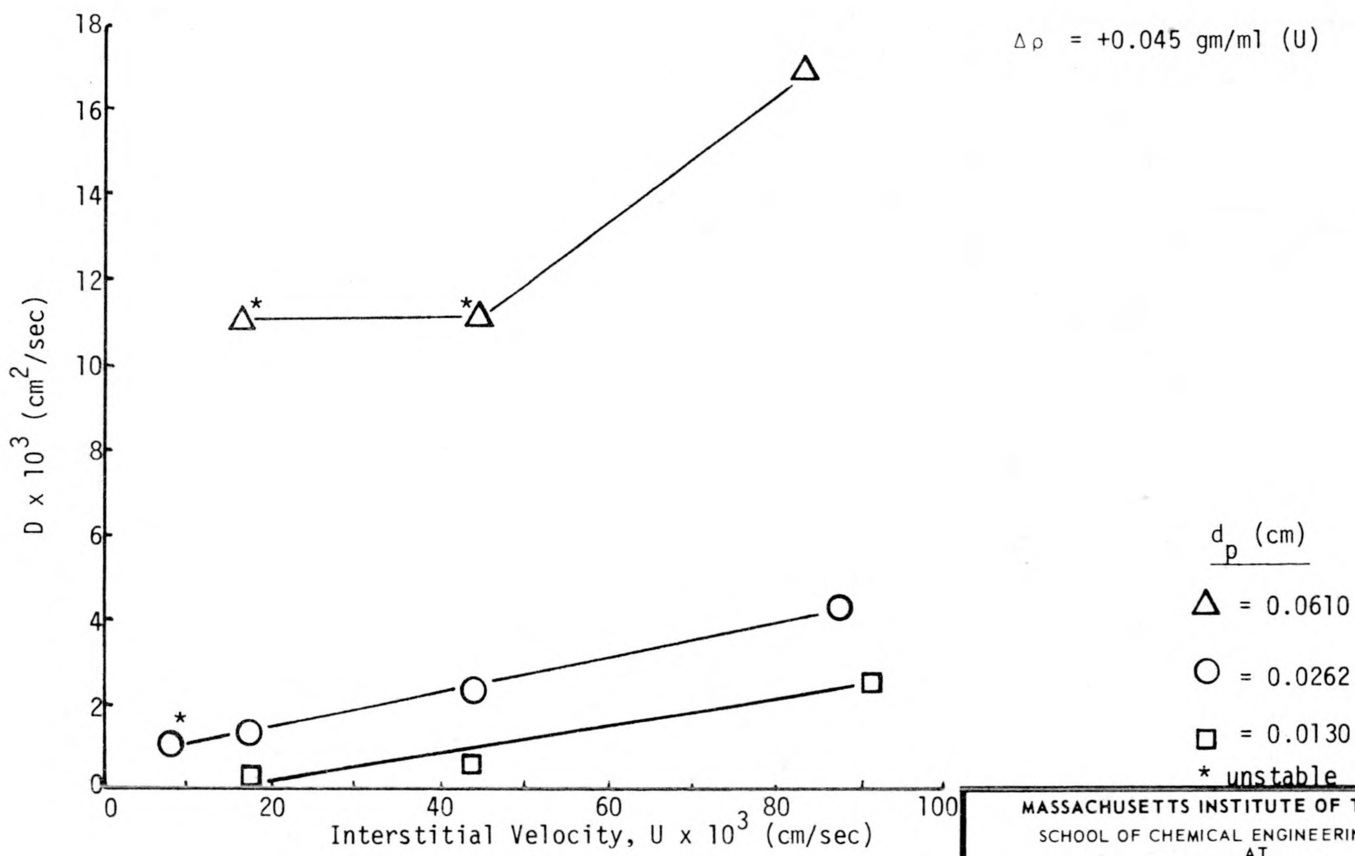

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DISPERSION COEFFICIENT
VS INTERSTITIAL VELOCITY

DATE	DRAWN BY	FILE NO.	FIG.
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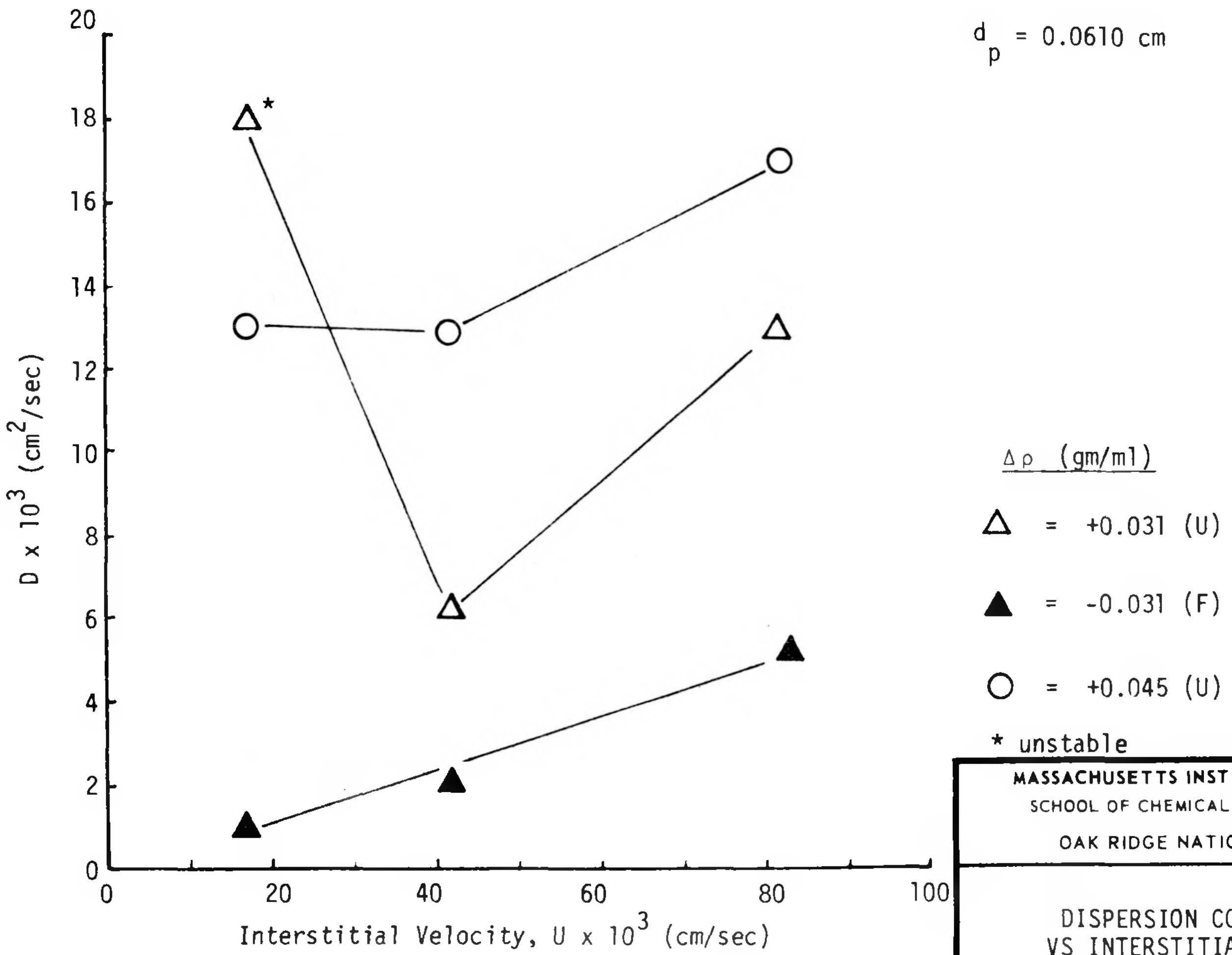

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DISPERSION COEFFICIENT
VS INTERSTITIAL VELOCITY

DATE	DRAWN BY	FILE NO.	FIG.
10-17-77	BAB	CEPS-X-259	12

4. The two-parameter dispersion model was found to describe the system quite well, even though the parameter f was essentially unity.

6. RECOMMENDATIONS

1. Salt solutions of equal viscosities should be used in future experiments to permit investigation of larger density differences without interference of viscosity peaks.

2. Further experimentation should be conducted using density unfavorable systems for a thorough description of the transition zone.

7. ACKNOWLEDGMENTS

We would like to thank our consultants, S.Y. Shiao and K.A. Kraus, for their guidance and suggestions throughout our project. J.S. Johnson offered several helpful recommendations and,M.H. Lietzke's assistance was also greatly appreciated. G.W. Westley's help with our computer modeling proved invaluable.

8. APPENDIX

8.1 Mathematical Model

Miscible fluid displacement can be described with a dispersion model (2) as:

$$D \frac{\partial^2 C}{\partial x^2} - V \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \qquad (3)$$

with the following boundary conditions,

$$\text{at } x = 0 \qquad VC_o = VC - D \frac{\partial C}{\partial x} \qquad (4)$$

$$\text{and } x \to \infty \qquad C(x,t) \to C_o \qquad (5)$$

$$\text{at } t = 0.0 \qquad C(x,t) = C_o \qquad (6)$$

The solution of Eq. (3) with boundary conditions (4), (5), and (6) is

$$\frac{C}{C_o} = \frac{1}{2} \operatorname{erfc}\left(\frac{\sqrt{\gamma}}{2} \frac{1.0 - I}{\sqrt{I}}\right) - \frac{\sqrt{I}}{\sqrt{\pi\gamma}(1.0 + I)} e^{-\gamma(1.0 - I)^2/4I}\left(1 - 2 \frac{I}{1 + I}\right) \qquad (7)$$

where I and γ are dimensionless time and position, respectively.

8.2 Location of Original Data

The original data may be found in ORNL Databook A-8144-G, pp. 1-34, and in the calculation file on file at the MIT School of Chemical Engineering Practice, Bldg. 3001, ORNL.

8.3 Nomenclature

C	concentration of displacing fluid, gm/ml
C_o	initial concentration of displacing fluid, gm/ml
D	dispersion coefficient, cm^2/sec
d_p	particle diameter, cm
f	fraction of total pure volume occupied by mobile fluid
g	gravitational acceleration, 981 cm/sec^2
I	Vt/L, dimensionless pore volume or time
K	porous bed permeability, cm^2/sec
L	column length, cm
L_M	mixing length, defined as fraction of pore volumes discharged between $C = 5\%\ C_o$ and $C = 95\%\ C_o$, dimensionless
P	pressure, atm
t	time, sec
U	velocity, cm/sec; also denotes unfavorable velocity
U_c	critical velocity, cm/sec
V	flow rate, ml/min
x	longitudinal distance in column, cm
γ	Vx/D, dimensionless dispersion
ρ	density, gm/ml
μ	viscosity, gm/cm-sec

8.4 Literature

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