Study of Multiphase Flow Useful to Understanding Scaleup of Coal-Liquefaction Reators

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Technical Progress Report

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Harold N. Knickle

Department of Chemical Engineering University of Rhode Island Kingston, R.I. 02881 401-792-2678 or 2655

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#### MULTIPHASE FLOW TECHNICAL PROGRESS REPORT June 1, 1983 to August 31, 1983

#### I. HIGHLIGHTS

Gas holdup measurements for air and aqueous carboxy methyl cellulose solutions have been performed using the 13 inch acrylic column. The experimental data has been analyzed and is reported here. A 1/4 inch thick porous plate with 70 micrometer pores made of polyethylene and an acrylic sieve plate with 1/8 inch diameter holes were used as gas distributors. Holdup was measured at different axial positions and compared to total holdup measurements by summing. The gas velocity range extended over the bubble and bubble-slug patterns.

Based on the results of this study a number of conclusions may be drawn about the effect of non-newtonian liquids on two phase flow parameters:

- 1. Rheological properties can affect gas holdup in bubble columns. The way in which these properties affect gas holdup depends on the flow pattern the column is operating in. In the bubble flow pattern gas holdup increases with viscosity at a particular gas velocity. In the bubble-slug or heterogeneous flow pattern, gas holdup generally decreases with viscosity.
- Liquid velocity affects gas holdup in the bubble flow pattern.
   Increasing the liquid velocity reduces the gas holdup at any particular gas velocity. This effect is accentuated by increased viscosity.
- 3. The bubble to bubble-slug transition is dependent on viscosity. The transition gas velocity decreases with increasing gas viscosity. There is no effect of liquid velocity on this transition however in the range studied.

4. The effect of alcohol on two-phase flow parameters was extreme. Holdup in the bubble flow pattern was very high, often greater than 0.5 peaks. The bubble to bubble-slug transition occurred at higher gas velocities than in aqueous CMC solutions. There also appeared to be competing effects between alcohol and CMC concentrations.

### II. OBJECTIVES AND SIGNIFICANCE

There are three major objectives for this proposed study. These objectives are basic to the understanding needed to develop a rationale for scale up in bubble columns. This understanding is the key to improving our scientific and technical knowledge of the fundamental process involved in complex two and three phase flows.

These objectives are:

- to properly characterize two phase flow patterns in the region of interest that direct coal liquefaction reactors will be operated.
- 2. to characterize for viscous liquids, Newtonian and non-Newtonian, the flow pattern boundaries in the operating region of direct coal liquefaction reactors. The characterization would include both empirical and theoretical models.
- 3. to develop empirical expressions and models for the gas holdup in the flow regimes of interest. This objective would focus on non-Newtonian liquids that follow some elementary models for constitutive behavior.

The significance to the fossil energy program includes:

 Flow pattern prediction will aid in the design and scaleup of coal liquefaction reactors.

- Gas holdup and estimates of bubble diameter will be useful in predicting interfacial area.
- Hydrodynamic modelling of the bubble column will be useful in predicting backmixing and residence times.

#### III. FLUN PATTERNS

It has been observed that two phase flow occurs in one of several different patterns. Which pattern a pair of fluids are flowing in depends on the respective velocities and the physical properties of the fluids. For two phase concurrent vertical flow, six different patterns are possible but only two are important in direct coal liquefaction bubble columns. These include bubble or homogeneous flow and bubble-slug transitional flow or heterogeneous flow. Figure 1-1 illustrates the characteristics of each pattern and their relative position in terms of gas and liquid velocities.

The bubble flow pattern is realized at low gas and liquid velocities. This pattern is characterized by an unimodal distribution of bubble sizes and burbles which rise independently of one another. This type of flow is often termed homogeneous or pseudonomogeneous due to the uniform bubble size and the uniform distribution of bubbles in the column.

The bubble-slug pattern occurs when the gas velocity is increased to the point where the individual bubbles interact to a large extent. Bubble coalescence occurs in this pattern, forming a bimodal distribution of bubble sizes. The larger bubbles tend to rise in the center of the column, this causes the liquid to circulate in distinct cells inside the column. Backmixing is thus greatly increased in this pattern.

### IV. NON-NEWTONIAN TWO PHASE FLOW

The effect of non-Newtonian behavior of liquids on two phase flow parameters has only recently become of interest. As far back as 1964 however this area has been studied.

Initial studies of bubble shapes and motion while rising in non-Newtonian liquids was conducted by Astirita (3). He found that for all of the fluids studied bubbles with volumes over 0.1 cc behaved according to:

$$V_{+} = 25.0 V^{1/6}$$
  $V = volume$ 

He also found for highly elastic fluids a critical bubble diameter of approximately 0.1 cc where the terminal velocity jumped by a factor as large as six.

non-Newtonian fluids. The most popular fluid used in these studies was solutions of carboxymethyl cellulose in water. The types of gas distributors used were varied; Nakanoh and Yoshida (30) used a single oriface sparger in a 14.55 cm column. Franz et al. (14) measured gas holdup in a multistage column, stages were separated by perforated plates. Deckwer et al. (10) and Schumpe and Deckwer (36) measured gas holdup for a number of CMC solutions using both porous and perforated plates (10) and also used a flexible plate in some tests. Others who used porous plates included: Buchholz et al. (5). Table 2-1 summarizes gas holdup correlations in existing literature.

Shear rates in bubble columns were measured by Mishikawa et al. (31). They found the shear rate to be related to gas velocity by;

This relationship is important since shear rates cannot be calculated

directly for two phase mixtures. It was also limited to superficial gas velocities a pore 0.04M/S.

Interfacial area for non-Newtonian liquids in bubble columns were measured by several researchers. Schumss et al. (37) measured interfacial area for CMC solutions by both chemical and photograhic methods, and found wide descrepancies between the two, aeas found by photographic method increased much faster with gas velocity than by the chemical method. Buchholz et al. (6) measured interfacial area by the photographic method. He reported a trimodal distribution of bubble sizes under some conditions in CMC solutions.

#### V. EXPERIMENTAL

Eight different non-Newtonian fluids were studied in the thirteen inch column (see table 3-1). These included four carboxymethyl cellulose (CMC) solutions and four CMC and isopropyl alcohol (IPA) solutions. CMC was chosen for its pseudoplastic behavior (see Appendix II), its popularity for use in researching non-Newtonian fluids, its stability under shear and its non-hazardous nature.

The rheology of CMC solutions is pseudoplastic and can be characterized using the power model, i.e.:

### T=K/gc(dVx/dy)n

Solutions containing IPA were utilized to provide a variation in surface tension.

Flow curves and power law coefficients (Figure 3-2) were determined using a Haake RV-12 viscometer. Surface tension was measured using a bougnoy-ring tensiometer.

Two gas distributors were utilized in this study; a polyethylene porous plate and a sieve plate. The porous plate was one quarter inch thick and 12 inches in diameter with an average pore size of 70 microns. The sieve plate was a one eight inch acrylic plate with one eight inch holes on an one half inch triangular pitch.

Gas velocities were varied for each run between 0.003 and 0.012M/S. This range of gas velocities were studied to provide measurements well into the bubble-slug flow pattern (see Figure 3-3). The number of velocities studied within this range varied between eight and ten depending on the fluid.

Three liquid velocities were studied for each fluid; 0, 0.005 and 0.012M/S. For each fluid and distributor plate, runs were repeated for each of these liquid velocities.

Bubble size distributions were determined for each fluid and distributor plate as a function of gas velocity. Still picture photography was utilized to measure bubble sizes. A transparent millimeter scale was included in all photographs to facilitate measurement of bubble diameters.

Photographs were taken on all runs where the liquid velocity was  $0.012 \, \text{m/s.}$  Photographs were taken at the column wall  $0.3 \, \text{meters}$  above the distributor plate.

### VI. FLOW PATTERNS: EXPERIMENTAL RESULTS

The bubble to bubble-slug flow transition was observed for all of the fluids studied. This transition varied with both CMC and IPA concentrations. Transitions were only observed with the porous plate gas discributor.

In most cases the transition point could be observed visually. However, in some CMC-IPA solutions the change was so subtle, the point where bubble slug flow began could not be pinpointed. This was the case with two of the CMC-IPA solutions, 0.25% CMC and 0.5% CMC. solutions a gradual appearance of substantially larger bubbles (20mm as opposed to 105 mm) with increasing gas velocity occurred. But the point at which the flow pattern changed was not obvious. The IPA solution with 0.75%wt CMC showed a well defined transition from bubble to bubble-slug while the IPA solution showed a dependence on height in the column. Coelesence began at the top of the column and moved downward with increased gas velocity. The behavior above and below the transition point in the column was easy to distinguish. The two phase mixture at the top was highly turbulent and very large bubbles occurred with a short frequency. The mixture at the bottom of the column displayed a lot of backmixing but not the violent action observed at the top and no very large bubbles were observed in the bottom.

A phenomenon that hampered observation of flow pattern transition in the IPA solutions was the occurrence of foam in the column. In the bubble flow pattern foaming was extensive and with no liquid flow the foam was observed to overflow the column for as long as twenty minutes. Once the transition point was past, the foam broke down however. At increased liquid velocities foam was less of a problem as it was continuously discharged from the column. At higher CMC concentrations (0.75 and 1.0%) foam was also hindered somewhat. The 1.0% solution showed only a pronounced zone at the top of the column at low gas velocities below the transition. The 0.75% solution showed more foam, however, it was considerably less than that observed at lower concentrations.

Analysis of the gas holdup data revealed flow pattern transitions at for the most part the same gas velocities as the visual observations. For both CMC solutions with and without IPA a general trend was observed in the transition gas velocity and CMC concentration. For both types of solutions this trend followed:

### Vgs(t)= A-B(CMC conc.)

gas holdup were not well defined in most of the gas holdup data. These errors were estimated by considering the distance between adjacent data points as the range in which the peak may lay. These errors were depicted on Figure 4-1.

### VII. GAS HOLDUP: EXPERIMENTAL RESULTS

Gas holdup was measured as a function of height in the column and gas velocity. In general gas holdup was higher with the porous plate than with the sieve plate. The most predominent difference was observed in the bubble flow pattern. In the bubble-slug flow pattern the differences were small.

The effect of CMC concentration on gas holdup with the sieve plate was predictable in solutions of CMC and water alone (figure 4-2), holdup decreased with increased CMC concentration. Gas holdup for CMC solutions with IPA was unpredictable (figure 4-3), the 0.5% solution gave higher holdup values than all of the others including the 0.25% solution. The 0.25% and 0.75% had almost identical holdup curves for the zero liquid flow case.

Gas holdup data with the porous plate revealed the two flow patterns in which the column was operated, bubble flow and bubble-slug flow. Gas

holdup in the bubble flow pattern increased faster with gas velocity than in the bubble-slug pattern. See figures 4-4 and 4-5.

The effect of IPA on gas holdup was pronounced. Gas holdup was observed to exceed 50 percent at peaks in several solutions; 0.0%, 0.25%, and 0.5% CMC. These excessive gas holdups were possibly due to foam accumulation in the column. Foam was observed in the bubble column with all of the IPA solutions. The lower concentrations of CMC resulted in higher foam production.

The effect of CMC concentration on gas holdup was to retard it. Gas holdup increased in the bubble flow pattern with increased CMC concentration. The gas velocity at maximum gas holdup in the bubble flow pattern decreased with increased CMC concentration. In the bubble-slug flow pattern the differences were substantially less however, between various CMC solutions.

The effect of liquid velocity on gas holdup was to decrease it in the bubble flow pattern, (figures 4-7, 4-8 and 4-9). There was a negligible effect on holdup in the bubble-slug flow pattern. IPA solutions showed little change in the bubble flow pattern however, peak holdup was reduced with increased liquid velocity; probably due to the expulsion of foam from the column.

#### VIII Correlation of Results

Flow pattern transitions for both CMC solutions and IPA and CMC solutions were found to be inversely related to CMC concentration and thus related to apparent viscosity. Since the shear rate in the bubble flow pattern and in the bubble to bubble-slug transition had never been determined, (Wishikaura (30) determined shear rates for heterogeneous

flow) a pseudo apparent viscosity was used. This pseudo apparent viscosity was defined as:

$$\mathcal{L}$$
 app=KVgs<sup>n-1</sup> 4-1

The transition gas velocity for acqueous CMC solutions was found to fit:  $Vgs=0.0023 \mu app^{-0.69}$  4-2

The transition superficial gas velocity could then be determined by inserting equation 4-1 into 4-2 and rearranging:

$$LnVgs = -$$
 (0.69n+0.31)

The transition gas velocity for CMC and IPA solutions were fitted similarly this resulted in:

-0.306

and

$$1.671+0.306LnK$$

$$LnVgc(1) = _____ 4-5$$

$$0.306n+0.694$$

The calculated and observed transition gas velocities were plotted in figure 4-10.

Equations 4-3 and 4-5 indicate that the transition from bubble to bubble-slug flow is dependent on the rheological properties of the liquid. Prior studies for newtonian liquids indicated the transition to be independent of viscosity.

Gas holdup data for both aqueous CMC solutions and CMC and IPA solutions were fitted to equations of the form:

$$(c-n)$$

(B-K)

For aqueous CMC solutions holdup data in the bubble flow patterns for the porous plate distributor were correlated. The resulting equation was:

The standard deviation of the differences for this equation was 0.015, (figure 4-11).

This correlation it should be noted is limited to the bubble flow pattern. The maximum gas velocity in this flow pattern can be determined using equation 4-3.

Gas holdup data for CMC solutions with the sieve plate distributor were fitted to:

The standard deviation from this curve was found to be 0.011 (figure 4-12).

There is no apparent physical explanation to the form of these equations in that the addition of a constant to k makes no physical sense. Further apparent viscosity or shear stress can not be extracted from k and the velocity term in either equation. The form of the gas

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reduced bubble rise velocities. The sign on the sign on the velocity terms however does have physical implications. The sign on the flow index "a" is positive for the bubble flow pattern and negative for the bubble slug. This indicates a different set of forces or a different balance of forces acting on the bubbles in both patterns. In the bubble flow pattern gas holdup is proportional to Vgs<sup>2</sup>·8n, this indicates that holdup increases with shear stress. It is possible that increased shear stress retards bubble vibrations and oscilations thus reducing interactions with other bubbles, this combined with decreased bubble rise velocities would result in increased gas holdup up to the point where bubbles are bunched too close together and begin to interact anyway. This phenomenon was noted by Schumpe and Deckwer (37) and attributed to reduced bubble rise velocities.

Gas holdup data for IPA solutions could not be correlated to any models similar to the ones used for aqueous CMC solutions. This lack of fit may have been due to the complex nature of these solutions. It was apparent that there were competing effects from the CMC and IPA in solution. Gas holdup was maximized in the 0.5% CMC solution and minimized in the one percent CMC solution. This would suggest that CMC and IPA interact at the interface in some way.

### IX Comparison of Results with Literature:

Schumpe and Deckwer did not discuss flow pattern transitions and the effect of CMC concentration or rheological properties on the flow pattern transitions. They did however provide a flow map whose coordinates are gas velocity and apparent viscosity. This flow map indicated a decrease in the bubble to bubble-slug flow pattern transition gas velocity with increased apparent viscosity. This agreed with the present study.

The gas velocities at peak gas holdup in the bubble flow pattern reported by Schumpe and Deckwer were plotted with the data from this study as both CAC concentrations and the gas velocity predicted by equation (4-3), see figures 4-13 and 4-14. Schumpe and Deckwer's data exhibited behavior that varied from the present study shown in figure 4-13. Their velocity vs. concentration data appeared to fall on a curve while data from this study fell closer to a straight line. This was probably due to differences in rheological properties of the solutions used and the use of a different gas distributor; Schumpe and Deckwer used a sintered plate with an average pore diameter of 150mm. In figure 4-14 Schumpe and Deckwer's data was plotted against gas velocities predicted with equation 4-3 and their viscosity data. This plot showed a closer relationship between the two sets of data, Schumpe and Deckwer's values were however higher than predicted for the most part. This was possibly due to the different gas distributors.

The correlation developed for gas holdup in the bubble flow pattern, equation 4-6 was plotted for a 1.0% CMC solution with Schumpe and Deckwer's correlation for gas holdup in the bubble flow pattern for solution concentration greater than 0.8%, see figure 4-15. Gas holdup in the bubble flow pattern was found to be higher in this study than reported by Schumpe and Deckwer. This was probably again due to difference in the gas distributors. Insertion of "k" and "n" for Schumpe and Deckwer solutions into equation 4-6, predicted generally higher holdup than they observed, see figure 4-16.

Equation 4-7 was compared with Schumpe and Deckwer's correlation for holdup using a sieve plate and Godbole et als correlation for gas holdup, also with a sieve plate, see figure 4-15. All three curves showed good agreement up to a gas velocity of 0.04m/s. At velocities above 0.04m/s

the curves diverged, Schumpe and Deckwer increased faster and Godbole et als increased slower than equation 4-6. Godbole et als curve was much closer to equation 4-6.

#### X Recommendations:

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There is a need for further investigation of two-phase flow with non-newtonian liquids. A more extensive variety of liquid properties need to be studied to separate rheological properties from other physical chemical properties such as ionic strength, surface activity.

Shear rates need to be determined in the bubble flow pattern to help understand the effect of viscosity on gas holdup. Shear rates are only known for heterogeneous flow at gas velocities above 0.04m/s.

Fast and reliable methods for determining interfacial area and bubble sized distributions need to be developed. Photographic techniques are tedious, photographs are often difficult to interpert and interpertations are often dependent on the individual. Chemical methods for determination of interfacial area provide good average values however, to axial and radial dependencies local methods are required.

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Table 1 Liquid Physical Properties

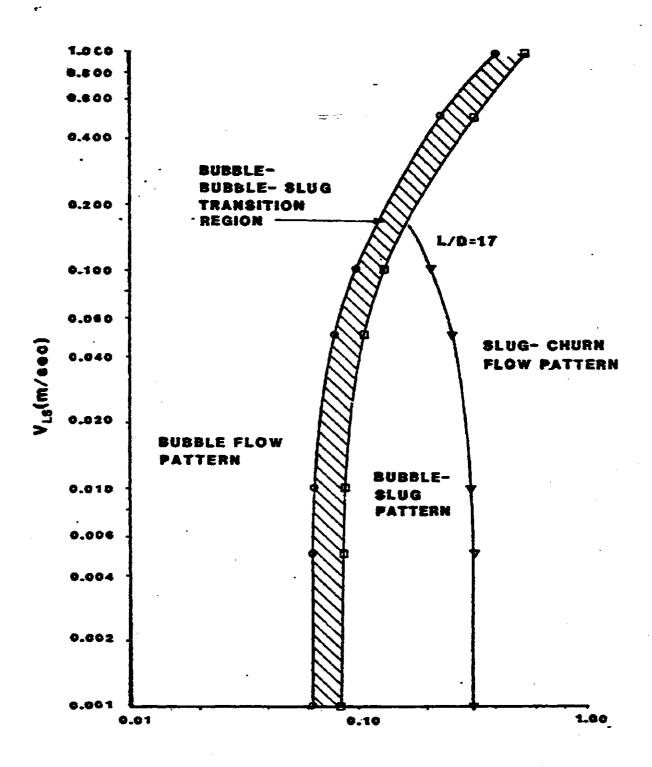
OMC Conc. (wt. %)	IPA Conc. (wt. %)	Specific Gravity	Surface Tension (Dynes/	Power Law K	Parameters N
0.0	0.0		CM) 72	0-01	1.0
0.25	0.0		72	0-012	0.97
0.5	0_0		72	0.15	0.95
0.75	0.0		72	0.025	0.91
1.0	0.0		72	0.04	0.88
0.0	8.0		45	0.009	1.0
0.25	8.0		45	0.018	0.958
0.5	8.0		45	0.034	0.918
0.75	8.0		45	0.06	0.878
1.0	8.0		45	0.095	0.833

# Table 2 Existing Holdup Correlations for Non-Newtonian Fluids

No.	Author	Correlations	Restrictions	References
1.	Alakansh & Yishida	= 0.20	Single Orifice Spargers	
2.	Godbole et al.	EG = 0.225 VBs532 Map. 146 CMC	Solutions Sieve Plate Distributors	
		EG = 0.0908 UGS 0.85	CMC conc 0.8% Sinteral Plates, Bubble Flow	
4.	Deckwer	E <sub>G</sub> = 0.0265 UBS82	Sieve Plates or Sintered Plate in Bubble-Slug Transition	

Table 3 Experimental Runs Using Aqueous CMC Solution

P	orous Plate		Sieve Plate				
Run#	CMC Conc.	IPA Conc. (wt. %)	Liquid Vel- (M/S)	Run#	OMC Conc. (wt. %)	IPA Conc. (wt. %)	Liquid Vel- (M/S)
2 3 4 5 12 13 14 18 19 20 21 22 30 31 32 33 34 44 45 46 50 51 52 56 57 58	0.0 0.5 0.5 0.25 0.25 0.25 1.0 1.0 0.75 0.25 0.25 0.25 0.5 0.5 0.75 0.75 0.75 0.75 0.75 0.75 0.75 0.75	0.0 8.0 8.0 8.0 8.0 8.0 8.0 0.0 0.0 0.0	0.0 0.005 0.012 0.005 0.012 0.005 0.005 0.005 0.005 0.012 0.0 0.005 0.012 0.0 0.005 0.012 0.0 0.005 0.012 0.0 0.005 0.012 0.0 0.005	1 6 7 8 9 10 11 15 16 17 23 24 25 26 27 28 35 36 37 38 39 40 47 48 55 55	0.0 0.5 0.5 0.25 0.25 0.25 1.0 1.0 0.75 0.75 0.25 0.25 0.5 0.5 0.75 0.75 0.75 0.75	0.0 8.0 8.0 8.0 8.0 8.0 8.0 8.0 8.0 0.0 0	0.0 0.0 0.005 0.012 0.0 0.005 0.012 0.0 0.005 0.02 0.005 0.12 0.0 0.005 0.12 0.0 0.005 0.012 0.0 0.005 0.012



V<sub>GS</sub>(m/sec) FLOW MAP FOR 0.333 METER COLUMN AIR- WATER SYSTEM (20°C, 1 ATM.)

Figure 1-1

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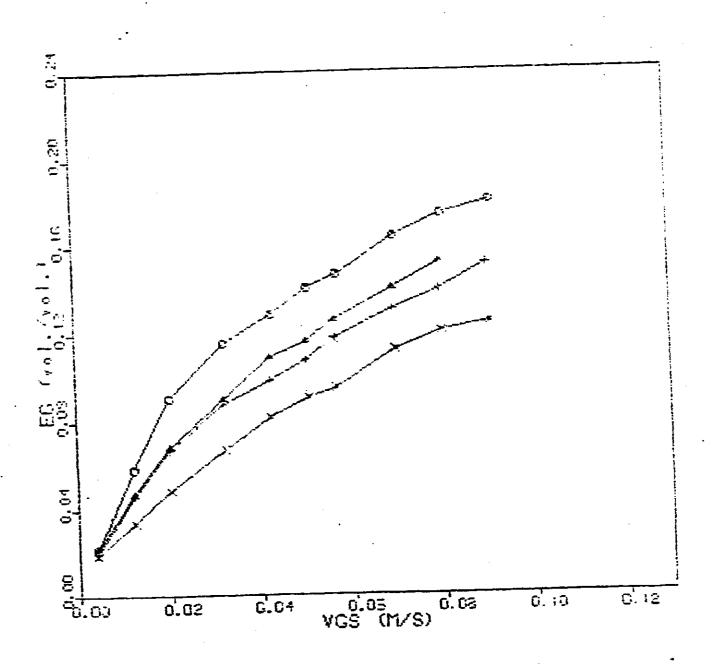
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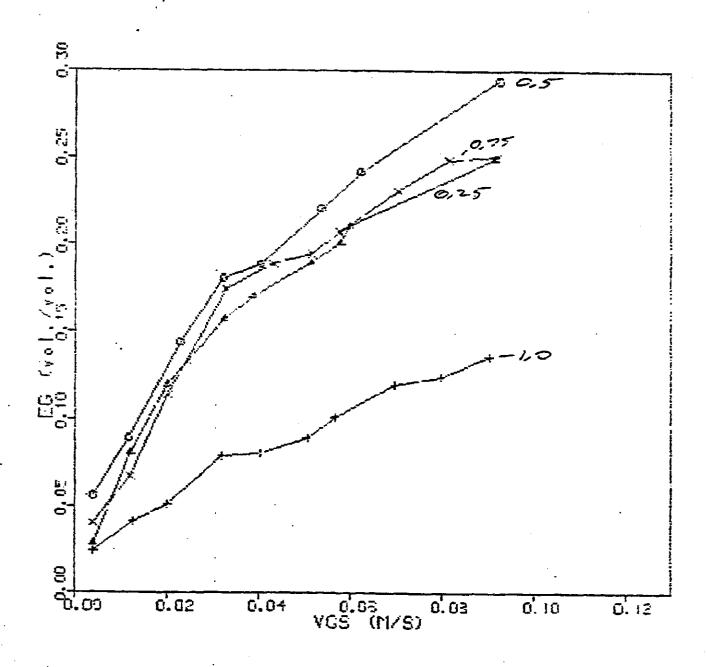
Sommous to tech Figure 4-1 Transition Gas Valocity us CMC Conc.

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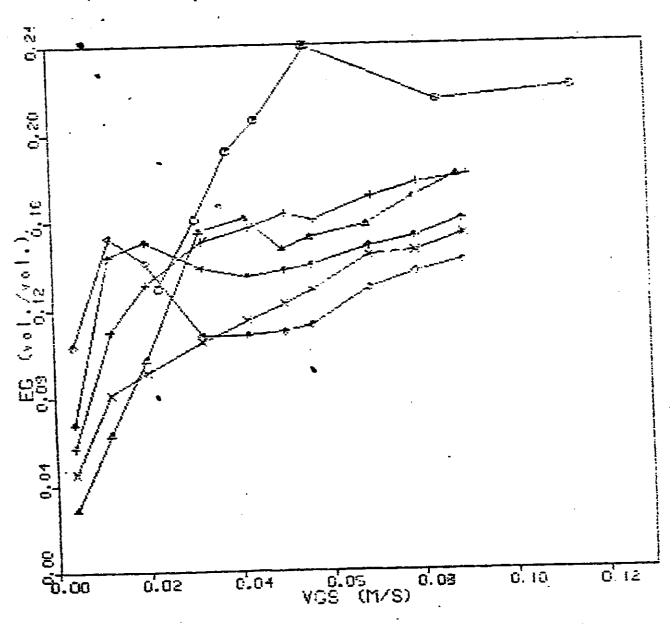
France 4-2

SYNSDL	CME CONC.	API CONO.	LIGHTO VELOCITY	DISTRIBUTOR
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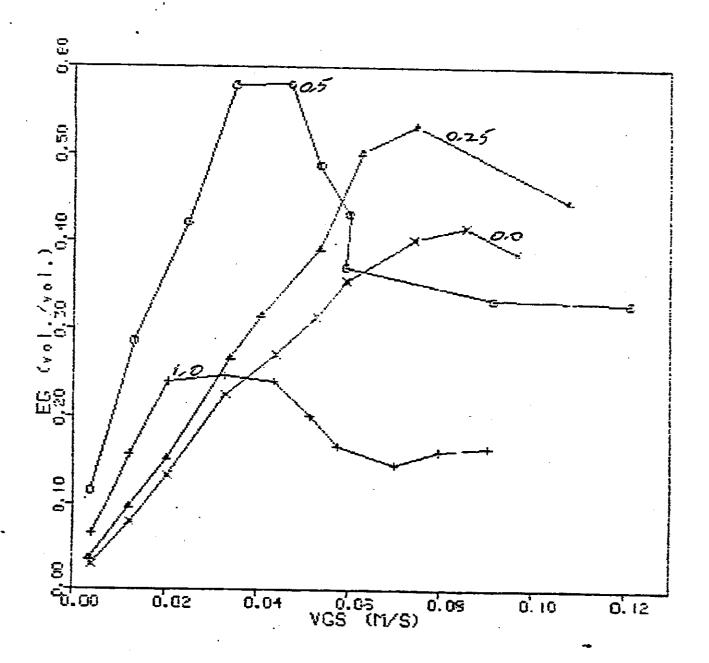
E -- 2 4-3

SYMBDL CMC CDNC. API CGNE. L1  6 0.0 .0  4 0.23 0.0  4 0.73 0.0  8 1.0 00  1 0.75 0.0	0 0 POROU PO
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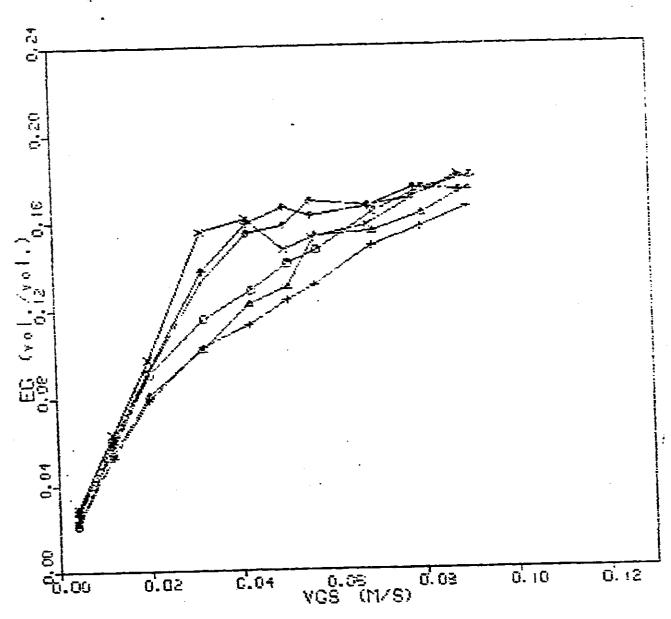
France 4-4

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+	10	80	0 012	PORGU
×	0.0	&, <u>Q</u>	0.012	POROJ



France 4-5

SYMBOL CMC CDMC. API COMC.  0 0.25 0.0  1 0.25 0.0  2 0.25 0.0  2 0.25 0.0  3 0.25 0.0  4 0.25 0.0	LECEND LIGHID VELOCITY 0.0 0.005 0.0 0.0 0.005 0.012	DISTRIBUTOR SIEVE SIEVE SIEVE POROU POROU
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	•		LEGEND	
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+	1 0	0 0	012	PGROU
×	1.0	0.0	0.8	SIEVE
	. 1.0	0.0	O. 005	5 (EVE
+	. 1.ບົ	0.0	Ö. 812	SIEVE

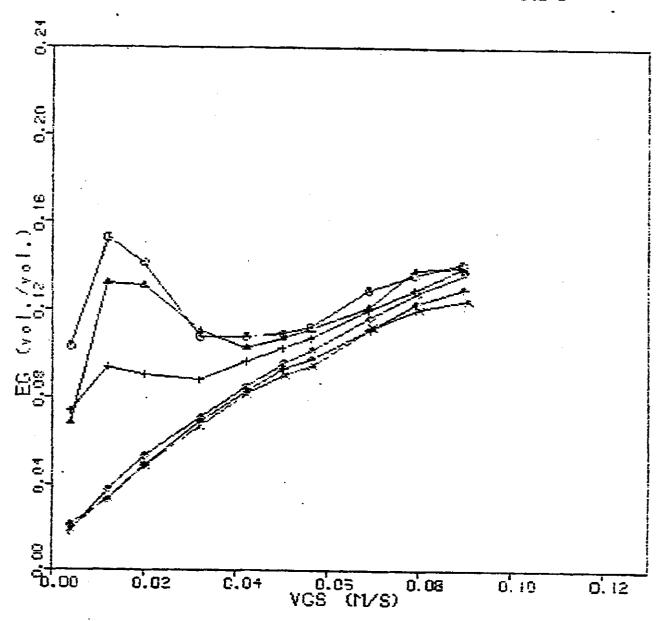
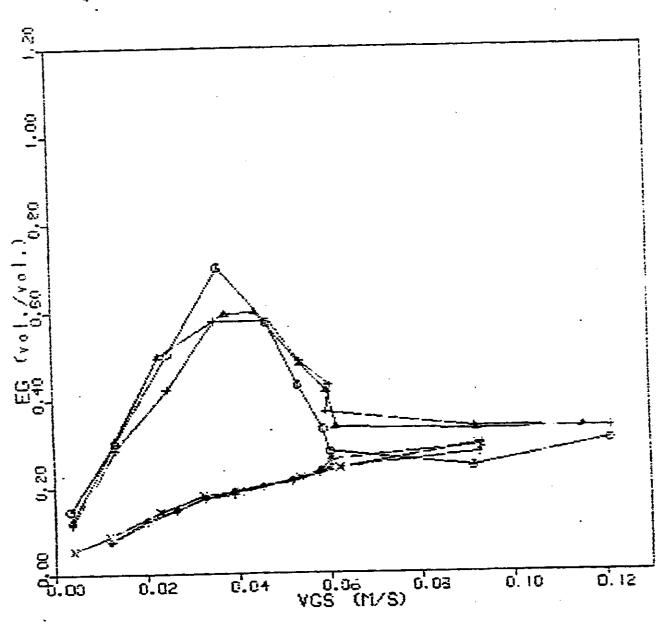


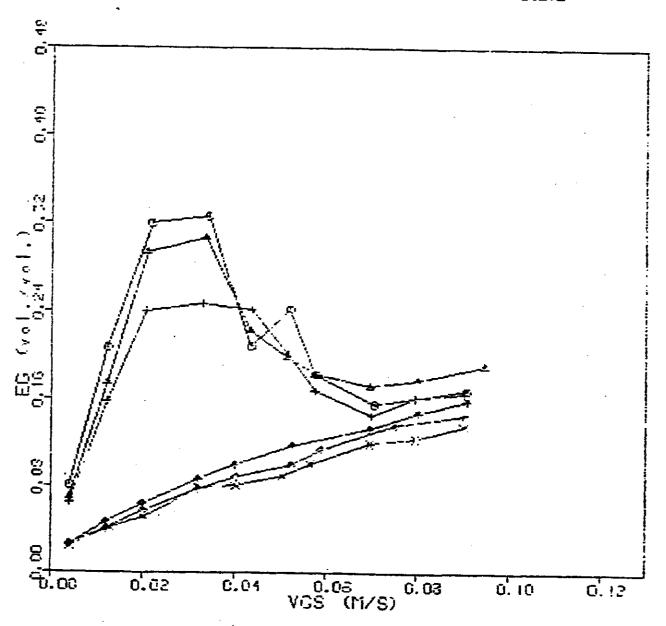
Figure 4-7

SYMBDL 6 4 + X	CHC CDMC. 0.5 0.5 0.5 0.5 0.5	.0 .0 & 0 &.0 <b>&amp;</b> .0	LFGERB LIQUED VELOCITY 0.0 0.005 0.005	DISTRIBUTOR POROJ POROJ FOROJ STEVE STEVE
€	6.5	8.0	G. 012	SIEVE



Fregue 4-3

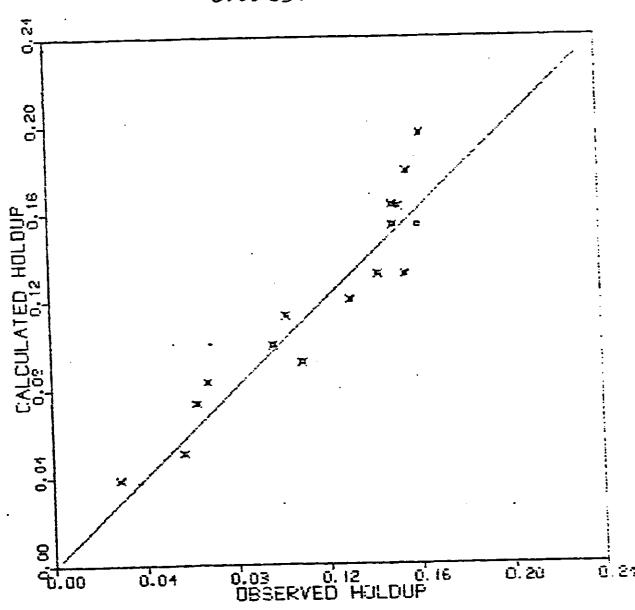
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+	1 0	ខេច	0 812	POROJ
>	1.0	8.0	0. G	SIEVE
•	. 1.0	S. 0	0.625	EIEVE
•	1.0	8.0	0.012	SIEVE



Fre - 21-0

## CALCULATED VS. OBSERVED HOLDUP

Model  $E_G = \frac{0.0694}{0.0583 + k} V_{GS}$ (n-0.761)2.8



## CALCULATED VS. OBSERVED HOLDUP

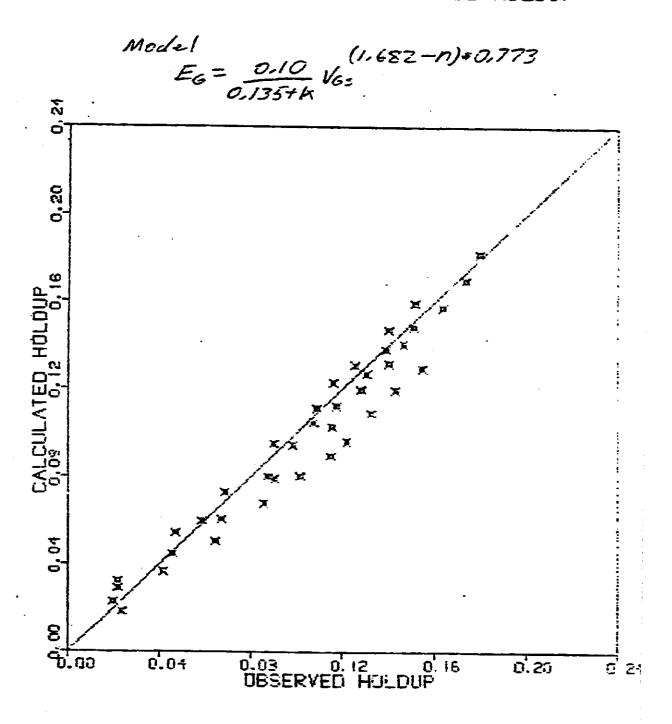


Fig., 2 4-12

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Figz: ~ 4-13

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Squares to the Inch

Figury 4-14

COMPARISON OF HOLDUP CORRELATIONS
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## DALCULATED VS. OBSERVED HOLDUP

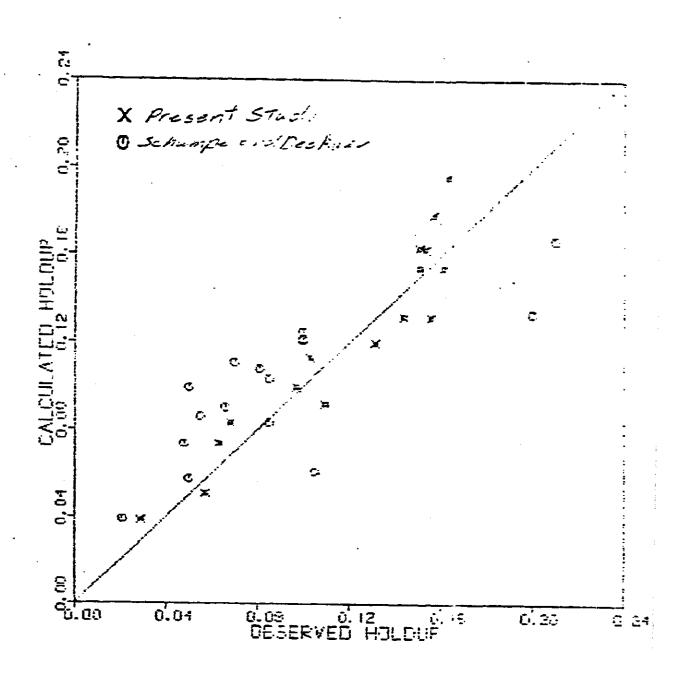


Figure 4-16