Study of Multiphase Flow Useful to
Understanding Scaleup of Coal
Liquefaction Reactors

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MULTIPHASE FLOW
TECHNICAL PROGRESS REPORT
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I. HIGHLIGHTS

The 13 inch acrylic column has been assembled. Tests using 20 different porous plates were performed on the six inch column, using air and water. Two non-Newtonian fluid have been selected for initial experiments. These pseudoplastic fluids will be a solution of carboxymethylcellulose and water and a solution of polyethylene oxide and water. A preliminary literature review of measurements with these solutions has been made. Properties to define these solutions including apparent viscosity, surface tension, and density have been identified and techniques to measure these properties are being evaluated.

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:

1. 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:

1. Flow pattern prediction will aid in the design and scaleup of coal liquefaction reactors.
2. Gas holdup and estimates of bubble diameter will be useful in predicting interfacial area.
3. Hydrodynamic modelling of the bubble column will be useful in predicting backmixing and residence times.

III. EQUIPMENT

A. 6 INCH ID COLUMN

A six inch inside diameter clear acrylic column with an overall height of about twenty feet has been used in this research program. Axial and total holdup has been measured in the column. Figure 1 is a schematic of the 6 inch system. A list of the characteristics of porous plate gas distributors for which gas holdup has been measured is presented in Table 1. These were used to study the effect of the gas distributor on holdup and the flow transition from bubble to bubble slug.

B. 13 INCH ID COLUMN

A thirteen inch inside diameter column of clear acrylic with an overall height of about twenty feet has been constructed. The column, structure, flow meters, centrifugal pumps, tanks, pressure regulators, control valves, filters, tubing, and gas distributors, have been installed. The superstructure has been installed and calibrated. Initial tests have been made with air and water. These will be compared with literature values of holdup. A schematic of the column is presented in Figure 2.

IV. SELECTION OF A NON-NEWTONIAN FLUID

Carboxymethylcellulose

Non-Newtonian liquids for initial measurements of holdup in both columns have been selected. One is a solution of a polymer, carboxymethylcellulose in water. Concentrations of CMC to be used in the first set of experiments will be 0.5, 1.0 and 2.0 weight %. These aqueous solutions are pseudoplastic and can be described either with the power law model

$$\tau_{yx} = -a \left| \frac{dv_x}{dy} \right|^{n-1} \frac{dv_x}{dy} \quad (1)$$

or with the Ellis model

$$\frac{dv_x}{dy} = (a + b |\tau_{yx}|^{n-1}) \tau_{yx} \quad (2)$$

The parameters for these models can be found in the literature and are presented in Tables 2 and 3. Schumpe et al (1) found that holdup decreases with concentrations of CMC greater than 0.8 wt %. They used a porous plate distributor and two bubble columns, 4.14 and 10. cm in diameter. Their gas holdup variation in the bubble flow pattern is given by the following equation

$$E_g = \text{const. } U_{GO}^{0.85} \quad (3)$$

where U_{GO} is the superficial gas velocity at the entrance of the bubble column. The gas holdup in the bubble-slug pattern is given by

$$E_g = \text{const. } U_{GO}^{0.67} \quad (4)$$

They (1) also found that the effect of the sparger in the bubble slug pattern is insignificant while the effect in the bubble pattern is significant.

Nicodemo et al (2) measured shear stress versus shear rate for a 3.0 wt% CMC solution at 23°C and determined the apparent viscosity with the power law model. They found the apparent viscosity varies with pH. Buchholz et al (3) also measured shear stress versus shear rate for varying CMC solutions up to 2.4 wt % and have determined the apparent viscosity for these solutions at 25°C. They also measured gas holdup in a bubble column 0.14 m in diameter and 3.91 m high. They found that gas holdup decreased with increasing concentration and reached a minimum at about 1.7 wt% CMC. This is essentially in agreement with Schumpe and Deckwer (1) even though the apparent viscosity is quite different.

Nishikawa et al (4) also determined the apparent viscosity of aqueous CMC solutions at 60°C and measured average shear rate versus superficial velocity. This last technique will be useful in deriving theoretical equations for flow pattern transitions.

These researchers indicate: (1) maximum gas holdup occurs at concentrations up to about 1 wt % CMC solution and, (2) the apparent viscosity is quite different for different samples of CMC.

Figure 3 is a graphical representation of the data given in Table 2 and shows apparent viscosity versus shear rate. Figure 4 is data redrawn for 0.5, 1.0, and 2.0 wt% CMC aqueous solutions. These are the solutions to be used in the first non-Newtonian hydrodynamic experiments. The densities of these solutions is presented in Figure 6.

It is clear that apparent viscosity, density and surface tension must be measured for each CMC solution.

Polyethylene Oxide

An aqueous polymer solution of polyethylene oxide will also be used in the holdup and flow pattern experiments. The polyethylene oxide has been purchased from Union Carbide. Table 4 is a list of the manufacturers data for two resins of different molecular weights. It is expected that concentrations of these polymers will be chosen for the apparent viscosity of the solutions less than 50 cp.

Previous measurements of the rheometric properties of these polymer solutions, Table 5, show that the solution is pseudoplastic. The exact values of these parameters will depend on the type of polymer used, i.e. high or low molecular weight. Measurements of apparent viscosity taken in our laboratory using a Brookfield viscometer are presented in Figure 7. Measurements taken by us using a Hakke rheometer are presented in Figure 8.

V. EXPERIMENTAL SET UP-DESCRIPTION

The schematic diagram of the experimental setup for the 6" column is depicted in Fig. 1. The bubble column is constructed of plexiglas and its inside diameter is 0.1524m(6") with a wall thickness of 0.0127m(1/2"). The total height of the column is 5.32m, consisting of four 1.33m sections flanged together. The gas distribution system occupies 0.1524m of the column, making the effective height of the column equal to 5.17m. A total of eleven pressure taps are in the column, the lowest being 0.1016m(4") above the distributor. The distances between the bottom five taps is 0.3048(12") and the remaining pressure taps are 0.6096m(24") apart. This enables the study of local holdup. The

pressure taps have been drilled with great care to reduce possible velocity head effects. A 0.8mm(1/32") hole has been drilled in the column wall and the inner surface polished to remove any burrs (eliminate velocity and friction heat effects). The tappings are all connected to a manometer board by means of flexible polypropylene tubing. The manometric board consists of 20 valves connected to a glass U-tube manometers filled with a manometric fluid. The connecting lines are filled with the column liquid.

Air is supplied by an existing compressor through a pressure regulator, air filter, rotameter, and a control valve to regulate the gas flow rate. Tap water is pumped from a 0.3785m (100 gallon) tank, through a water filter, control valve and a rotameter. Air enters the column at the bottom, through the gas distributing region. Three different positions for the liquid entrance directly to the column are located 0.0508m(2"), 0.06096m(24") and 1.371m(54") above the distributor plate. The air-water mixture flows up the column, and then into a gas-liquid separation tank, where the air is vented and the water recirculated back to the storage tank. Quick closing ball valves are attached to the entrance (both air and water) and exit section of the column. The air rotameter has been calibrated using an air flow meter, while the water rotameter has been calibrated by collecting the water in a tank for a specific time and measuring its volume.

The gas distribution system, consists of a conical section packed with 3.5mm borosilicate glass beads, and a 0.127m(5") diameter distributor plate placed on top. The distributor plate is fitted, so as to make sure air enters only through the distributor plate, and not through the side of the plate. Air entering from the sides, usually in the form of big bubbles, disrupts the flow regime, especially in the case

of uniform bubbling. Silicone seal is used to insure there are no leaks of air at the edge of the gas distributors.

Liquid and gas flow meters have been calibrated. The gas flowmeter was calibrated at atmospheric pressure. Since the column pressure at the entrance is greater than atmospheric the flow rate must be multiplied by the square root of the pressure ratio to determine the actual entrance flow rate.

VI. EXPERIMENTAL PROCEDURE

The experiment is started by filling the column with water, and flushing the connecting lines (to the manometers) to get rid of any existing air bubbles. The lines are then connected to the manometers. Before the air is bubbled through the column, there should be no pressure difference registered by the manometers, as the density of the column fluid and the connecting lines fluid are equal. After removing all air bubbles from the connecting lines, the air control valve is opened, and the air pressure regulator set to 30psig. The air and the water flow rates are adjusted to a desired value and the system is allowed to equilibrate for about 300 to 600s or when the manometer indicates a steady reading. The air and water flow rates along with manometer readings are noted. The temperature of the column is not controlled independently; thus a small temperature variation can take place during the course of the work. Since all the experiments will be performed inside it is expected that the temperature will vary between 20-25°C. The experiment will be repeated for different liquid flow rates and also for no liquid flow. At each liquid rate, measurements will be taken for a number of different gas rates. The gas and liquid flow rates will be

chosen, such that the superficial gas and liquid velocities are in the bubble and bubble-slug flow regime of the flowmap (Fig. 9). The flow patterns will be characterized by visual observations.

The equation for the transition line between the bubble flow pattern and the bubble-slug flow pattern is (5,6)

$$VLS = 2.2 VGS - 1.128 [g (\rho_l - \rho_g) \sigma / \rho_l^2]^{1/4} \quad (5)$$

To determine the transition from bubble to bubble-slug flow pattern the liquid properties, density and surface tension and the gas density must be known. An attempt will be made to modify this equation to viscous and non-Newtonian systems. Hydrodynamic studies of flow pattern and holdup will attempt to justify this equation as well as to determine the holdup at the transition.

VII REFERENCES

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2. Nicodemo, L., L. Nicolais, and R.F. Landel, "Shear Rate Dependent Viscosity of Suspensions in Newtonian and Non-Newtonian Liquids" Chem. Eng. Sci., 29, pp 729-735 (1974).
3. Buchholz, H., R. Buchholz, J. Lucke, and K. Schugerl, "Bubble Swarm Behavior and Gas Adsorption in Non-Newtonian Fluids in Sparged Columns," Chem. Eng. Sci., 33, pp 1061-1070 (1978).
4. Nishikawa, M., H. Kato, and K. Hashimoto, "Heat Transfer in Aerated Tower Filled With Non Newtonian Liquid," Ind. Eng. Chem. Process Des. Dev., 16, p. 133 (1977).
5. Knickle, H.N., and A.C. Kirpekar, "A Proposed Flow Map for Use in the Design of Coal Liquefaction Bubble Column Reactors," paper presented at AEChE Meeting, Orlando, (1982).
6. Knickle, H.N., and Kamat, A.B., "Prediction of Hydrodynamic Effects in Bubble Column Type Coal Liquefaction Reactors" to be presented at AIChE Meeting, Los Angeles, (1982).

TABLE I

POROUS PLATE DISTRIBUTIONS

Gas Distr.	Key	Pore size	Thickness	Material	Characteristics
1	PEL1	35u	1/4" = 0.00635m	Polyethylene	Hydrophilic
2	PEL2	35u	1/2" = 0.0127m	Polyethylene	Hydrophilic
3	PEL3	70u	1/16" = 0.0015875m	Polyethylene	Hydrophilic
4	PEL4	70u	1/8" = 0.003175m	Polyethylene	Hydrophilic
5	PEL5	70u	1/4"	Polypropylene	Hydrophilic
6	PEL6	70u	1/2"	Polypropylene	Hydrophilic
7	PEL7	70u	3/4" = 0.01905m	Polyethylene	Hydrophilic
8	PPL1	125u	1/4"	Polyethylene	Hydrophilic
9	PPL2	125u	1/2"	Polyethylene	Hydrophilic
10	PEB1	70u	1/16"	Polyethylene	Hydrophobic
11	PEB2	70u	1/8"	Polyethylene	Hydrophobic
12	PEB3	70u	1/4"	Polyethylene	Hydrophobic
13	PEB4	70u	1/2"	Polyethylene	Hydrophobic
14	PEB5	70u	3/4"	Polyethylene	Hydrophobic
15	SC1		1/8"	Silicon Carbide	
16	SC2		1/4"	Silicon Carbide	
17	A01	.062" 1" = 0.025 m (1.575x10 ⁻³ m)		Aluminum oxide	34 to 38% porous Density = 2.2 to 2.3 lb/in ³
18	A02	.104" 1" (2.642x10 ⁻³ m)		Aluminum oxide	34 to 38% porous Density = 2.2 to 2.3 lb/in ³
19	A03	240u	1"	Fused Alumina grains	Permeability 50 cfm/ft ²
20	A03	240u	1"	Fused Alumina grains	Permeability 19 cfm/ft ²
21	CU1	30u	1/2"	Copper	
22	ZR1		1/2"	Zirconium Oxide	

TABLE 2
Power Model Parameters For Aqueous Solutions of CMC⁽¹⁾
At Room Temperature⁽²⁾

Composition (weight %)	a ($\text{lb}_f\text{-sec}^n\text{-ft}^{-2}$)	n (dimension less)
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(1) Carboxymethylcellulose

(2) Metzner, A. B., Advances in Chemical Engineering,
Vol. I, Academic Press, New York (1956) p. 103.

TABLE 3
Ellis Parameters For Aqueous
Solutions of CMC AT 85°F(1,2)

Composition (Weight %)	Molecular Weight	a (cm ² /sec-dyne)	b (cm ²ⁿ /sec-dyne ⁿ)	n (dimensionless)	Experimental Range of Shear Stress (dyne/cm ²)
4.0	Low	0.1377	0.3211	1.170	8 to 440
5.0	Low	0.00	0.0521	1.337	8 to 1010
1.5	Medium	0.4210	0.2724	1.185	6 to 300
2.5	Medium	0.0383	0.0181	1.412	17 to 1720
0.6	High	0.2891	0.0280	1.707	8 to 270

(1) Slatery, J. C., Doctoral Thesis, University of Wisconsin (1959), p. 79a.

(2) Bird, R. B., Stewart, W. E., and Lightfoot, E. N., Transport Phenomena, John Wiley and Sons, Inc., New York, (1960) p. 13.

TABLE 4
Union Carbide Polyox-Polyethylene Oxides
Water Soluble Resin⁽¹⁾

<u>Grade</u>	<u>Approximate Molecular Weight</u>	<u>Viscosity 25°C, cp 5% Aqueous Solution</u>	<u>Brookfield Spinole No/ Speed, rpm</u>
Polyox WSR N-10	100,000	12-50	1/50
Polyox WSR - 205	600,000	4500-8,800	2/2

(1) Polyox Water Soluble Resins - Union Carbide Manufacturers Literature (1981).

TABLE 5
Power Law Parameters For
Aqueous Solutions*

Solution	T(K)	$(N - S^n/m^2)$	$n(-)$
1.0 % Polyethylene Oxide (pseudo plastic)	293	0.994	0.532
	313	0.706	0.544
	333	0.486	0.599

*R. M. Turian, Ph.D. Thesis, University of Wisconsin, Madison (1964), pp. 42-148 in "Dynamics of Polymeric Fluids by R. B. Bird, R. C. Armstrong, and O. Hassager Volume 1, John Wiley & Sons (1977) p. 209

Schematic Diagram of Bubble column Set up.

FIGURE 2.

FIGURE 3

Apparent viscosity	vs	Shear Rate	for CMC.
			wt
I			0.67%
II			1.5%
III			3.0%

FIGURE 4

Apparent viscosity vs shear rate for CMC

I = 0.5% CMC (wt)
 II = 1.0% CMC
 III = 2.0% CMC

water $10^{-3} \text{ kg/(m}\cdot\text{s)}$

Density vs Concentration For CMC. at 8°C

FIGURE 6

K&E 10 X 10 TO THE INCH 46 0702
MADE IN U.S.A.
KEUFFEL & ESSER CO.

1020

1010

1000

990

980

0

1

2

3

4

5

WEIGHT % CMC

APPARENT VISCOSITY vs. SHEAR RATE for POLYOX WSRN-10

FIGURE 7 VISCOSITY CHARACTERISTICS OF AQUEOUS POLYETHYLENE OXIDE

ROTOVISCO RV-100 FLOW CURVE

25 % τ
80 Pa

HAAKE

$$\tau = A \cdot \% \tau \cdot S_\tau \text{ [Pa]}$$

$$D = M \cdot \% D \cdot S_D \text{ [s}^{-1}\text{]}$$

$$\eta = \tau / D \text{ [Pa} \cdot \text{s]}$$

Date 10/6/82

No. 1

Substance WCE
30

Temperature WSP
25°C

ROTOVISCO RV 100

System M-500

Sensor system MV-1

Factor A 3.22

Factor M 11.9

Program times:

T₁ 0

T₂ 3

T₃ 0

Signature 10

65 % D
760 s⁻¹

FIGURE 8 POLYETHYLENE OXIDE AQUEOUS SOLUTION (3%)

FIG. 11.9

FLOWMAP WITH PROPOSED CORRELATION INDICATING ZONE OF TRANSITION - AIR - WATER SYSTEM, 22° C, 1 ATM^(1,2)

- 1) KNICKLE, & KAMAT Submitted For Presentation at the Los Angeles AIChE Meeting, November 1982
- 2) KNICKLE & KIRPEKAR Presented at the Orlando AIChE Meeting, March 1982