TO UNDERSTANDING SCALEUP OF

COAL LIQUEFACTION REACTORS

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#### SUMMARY

Research over a three year time span involved the study of multiphase flow useful to understanding the scaleup of coal liquefaction reactors. We attempted to establish the flow patterns and their boundaries in which a direct coal liquefaction, large diameter, bubble column operates. A flow map has been proposed in which coal slurry properties can be input pattern boundaries flow at reactor determine the operating conditions. A new flow pattern had to be introduced, bubble-slug flow, because of the effect of the large diameter of these reactors. It has been established that the reactor would probably operate in the bubble or bubble-slug flow pattern.

Gas holdup and bubble diameters have been measured under different conditions of gas and liquid flow rate. These have been used to determine interfacial areas in bubble columns. An equation for the estimation of interfacial area in the bubble-slug flow pattern has been proposed. It has been established that physical methods are adequate to determine interfacial area in the bubble-slug pattern.

It has also been established that gas holdup and thus interfacial area depends strongly on the gas distribution in the column. Porous plate gas distributors can yield gas holdups twice as large as sieve plate distributors.

Measurements using carboxy methyl cellulose aqueous solutions indicate that non-Newtonian solutions can affect the gas holdup, the transition from bubble to bubble-slug flow, and the interfacial area. These experiments have been analyzed and equations for gas holdup and interfacial area have been proposed.

Analysis of these experiments involved modelling the bubble slug flow pattern with limited success. In highly viscous systems slug flow can reappear and modelling results for this pattern under highly viscous conditions were successful resulting in an equation for the shear rate in the bubble column. This last model also predicted gas holdup with reasonable accuracy.

Since these experiments were performed with two phase systems further research is suggested that would test the assumption of a homogeneous liquid solid mixture.

### RESULTS - DESIGN EQUATIONS

GAS HOLDUP: /NEWTONIAN LIQUID

Gas holdup correlations for air-water, systems in bubble columns are given below.

For a hydrophilic porous plate gas distributor for the air-water system gas holdup can be estimated from the following three equations:

In bubble flow

$$E_{c} = 7.128 \text{ (VGS)}^{0.824} \text{ (Thic)}^{0.102} \text{(Pore)}^{0.0477}$$
 (1)

In bubble-slug flow

$$E_{\rm G} = 0.428 \, (\text{VGS})^{0.305}$$
 (2)

In the transition between bubble and bubble slug flow

$$E_G = 0.125 \text{ (VGS)}^{-0.332} \text{ (Thic)}^{0.099} \text{ (Pore)}^{-0.0386}$$
 (3)

For a hydrophobic porous plate gas distributor for the air-water system gas holdup can be estimated from the following three equations:

In bubble flow

$$E_G = 4.808 \, (VGS)^{0.93} (Thic)^{0.0436}$$
 (4)

In bubble-slug flow

$$E_{G} = 0.428 \text{ (VGS)}^{0.305}$$
 (5)

In the transition between bubble and bubble slug flow

$$E_{G} = 0.159 \text{ (VGS)}^{-0.565} \text{ (Thic)}^{0.165}$$
 (6)

For a sieve plate gas distributor for the air-water system in bubble and bubble-slug flow, gas holdup can be estimated from the equation

$$E_{G} = 0.7008 \, (VGS)^{0.558} \tag{7}$$

GAS HOLDUP: NON NEWTONIAN LIQUID

For air-non Newtonian liquid (CMC) systems gas holdup is a function of apparent viscosity of the non Newtonian liquid.

For air-non Newtonian liquid system in bubble-flow gas holdup can be estimated from the equation

$$E_G = 3.52 (VGS)^{0.79} (\mu_{app})^{0.09}$$
 (8)

Gas holdup in bubble-slug flow can be estimated from the equation

$$E_{\rm G} = 0.159 \, (VGS)^{0.48} \, (\mu_{\rm app})^{-0.25}$$
 (9)

Gas holdup in slug flow can be estimated from the equation

$$E_{G} = 0.415 \, (VGS)^{0.66} \, (\mu_{app})^{-0.09}$$
 (10)

#### GAS HOLDUP: ENTRANCE EFFECT

Gas holdup measurements in bubble columns indicate an entrance region of about one meter and no column diameter effect between 6 and 13 inches.

## TRANSITION VELOCITIES: NON NEWTONIAN LIQUID

The transition superficial gas velocity from bubble to bubble slug flow, for non Newtonian systems (CMC solutions) can be estimated from the equation

$$VGS = 0.0023 \ (\mu_{app})^{-0.61} \tag{11}$$

The transition superficial gas velocity from bubble to bubble slug flow, for CMC and IPA solutions, low surface tension, can be estimated from the equation

$$VGS = 0.188 \ (\mu_{app})^{0.306} \tag{12}$$

#### SHEAR RATE: NON NEWTONIAN LIQUID

The average shear rate of the liquid phase in the column in the slugflow pattern can be calculated from the equation

$$\dot{\gamma} = \frac{2LTB (e_g/K)^{1/n} \left(r_0 - r_f\right)^{1/n + 2} + r_f(r_0 - r_f)^{1/n + 1} + 1.15(LLS)(ULLS)(r_0)}{LTB (r_0^2 - r_f^2) + r_0^2 LLS (1 - \alpha_{LS})}$$

#### BUBBLE SIZE ESTIMATION:

In the bubble pattern there is a unimodal distribution with average bubble sizes estimated for air-water system to be about 3mm. In the bubble-slug pattern there is a bimodal distribution with bubble sizes of about 3mm for the smaller bubbles and 7.5 to 12mm for the larger bubbles.

Radial distribution of the smaller bubble sizes at different axial positions were estimated for different concentrations of CMC solution. The following table illustrates the variability of the average radial distribution value of the smaller bubble size with increasing viscosity.

% CMC	Average Smaller Bubble Size	
0.0	3mm	
0.25	3.225	
0.5	3,58	
1.0	3.34	
1.5	2.6825	
2.0	2.5225	

Although the smaller bubble size decreases slightly with increasing viscosity it is recommended that the value of 3mm be used as an approximate value in design calculations.

#### INTERFACIAL AREA ESTIMATION:

The general form of interfacial area in bubble flow, where unimodal distribution of bubble size occurs, is given by the equation

$$a = 6E_G/d_R$$

and in bubble-slug flow, where bimodal distribution of bubble sizes occurs, interfacial area is given by the equation

$$a = \frac{6E_{G1}}{d_{B1}} + \frac{6E_{G2}}{d_{B2}} \frac{V_2}{V_1}$$

By using bubble sizes given previously and the results of gas holdup and dynamic gas disengagement experiments these equations can be transformed into design equations dependent only on gas holdup. Assume that the peak gas holdup observed in the bubble pattern exists in the bubble-slug pattern in the region surrounding the larger bubbles. This value of holdup represents

$$\frac{E_{G'}}{V_{T}-V_{LB}}$$

the small bubble volume fraction beyond which coalescence generates the larger bubbles of the bubble-slug pattern.

INTERFACIAL AREA: BUBBLE FLOW PATTERN

For a Newtonian fluid in bubble flow, interfacial area can be estimated from the equation

$$a = 20 E_{G} \tag{14}$$

Gas holdup to be used in the above equation can be estimated from equations (1), (4) or (7) depending on the gas distributor.

For a non Newtonian fluid in bubble flow, interfacial area can be estimated from the equation

$$a = 15 E_{G}$$
 (15)

Gas holdup to be used in the above equation can be estimated from equation (8) by knowing superficial gas velocity and apparent viscosity of the non-Newtonian liquid.

#### INTERFACIAL AREA: BUBBLE SLUG FLOW PATTERN

For a Newtonian fluid in bubble-slug flow, interfacial area can be estimated from the equation

$$a = 20 E_{G1} + 22.07 E_{G2}$$
  
( $d_{B1} = 0.3 cm; d_{B2} = 0.975 cm; = 3.587$ )

For the air-water system

$$\frac{V_{SB}}{V_T - V_{LB}} = 0.24$$

Substituting and rearranging the above equations, we get the design equation for interfacial area for a Newtonian fluid in bubble-slug flow as

$$a = 22.72 E_{G} - 0.65$$
 (16)

Gas holdup to be used in the above equation can be estimated from equations (2), (5) or (7) depending on the gas distributor.

For a non Newtonian fluid in bubble-slug flow, interfacial area can be estimated from the equation

$$a = 20 E_{G1} + 20.53 E_{G2}$$
  
( $d_{B1}=0.3cm$ ;  $d_{B2}=4.5cm$ ; = 15.4)

For a non-Newtonian fluid

$$\frac{V_{SB}}{V_T - V_{LB}} = 0.155$$

Substituting and rearranging the above equations, we get the design equation for interfacial area for a non-Newtonian fluid in bubble-slug flow as

$$a = 20.63 E_{G} - 0.1$$
 (17)

Gas holdup to be used in the above equation can be estimated from equation (9) by knowing superficial gas velocity and apparent viscosity of the non-Newtonian liquid.

COLUMN SIZING: DESIGN ALGORITHM

An accurate flow map will form the basis of any design calculations.

A design algorithm to size the reactor might be as follows:

- 1. Set the plant output (eg 100tons/day of coal oil)
- 2. Choose a column diameter (not a trivial decision)
- Calculate the gas, liquid and solid flow rates from material balances.
- Set the choice of the gas distributor (not a trivial decision)

- 5. Determine the flow pattern that will exist in the proposed reactor from the flow rates and typical physical property data.
- 6. Insure that the pilot plant runs with the best yields and is also operated in that flow pattern.
- 7. Set the residence times of the three phases in the proposed column from laboratory and pilot scale experiments (considering holdup and backmixing).
- 8. Calculate the height of the column necessary to obtain these residence times in the proposed column

This will assuredly be an iterative process.

#### RECOMMENDED FLOW MAPS:

A flow map is important in order to know the flow pattern and the flow transition of operation. These flow maps are recommended for large column diameters, greater than 0.1524 meters and heights greater than 1.5 meters. All the flow maps given show the transition from one flow pattern to the other. A design theorem for column scaleup requires the same flow pattern as the pilot plant to be used in all design calculations.

Figure 1 is a flow map for air-water system at 22°C and 1 atm pressure for a column diameter of 0.1524 m. It is a plot of VLS vs. VGS in m/s. This figure gives the various flow patterns that are observed in vertical two phase flow, and their transition boundaries. Knowing the superficial gas and liquid velocities from the experimental conditions, we will be able to estimate the flow pattern of operation and its transition boundary.

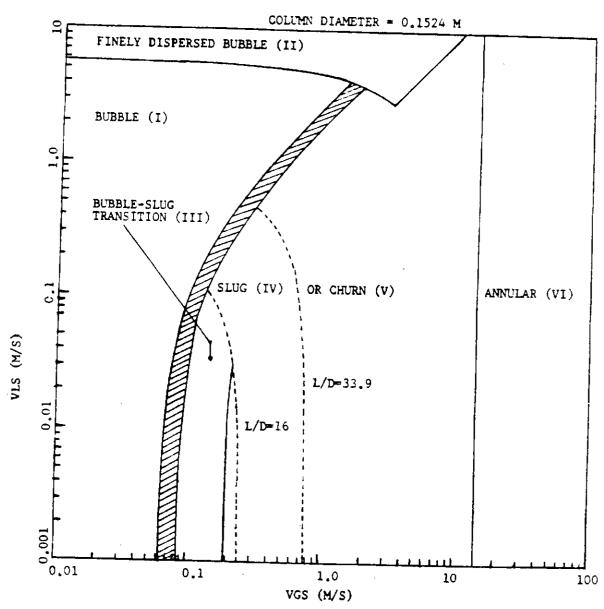


FIG. 1 MODIFIED. TAITEL ET AL FLOWMAP

AIR-WATER SYSTEM, 22 C, 1 ATM

Figure 2 is a flow map with VLS vs. VGS as the coordinates, for a 0.33655m diameter column for air-water system at 20°C. This figure is similar to Figure 1 but is applicable for a larger diameter, ie; 0.33655m diameter column. This figure gives the various flow patterns that are observed in vertical two phase flow, and their transition boundaries. Knowing the superficial gas and liquid velocities from the experimental conditions, we will be able to estimate the flow pattern of operation and its transition boundary.

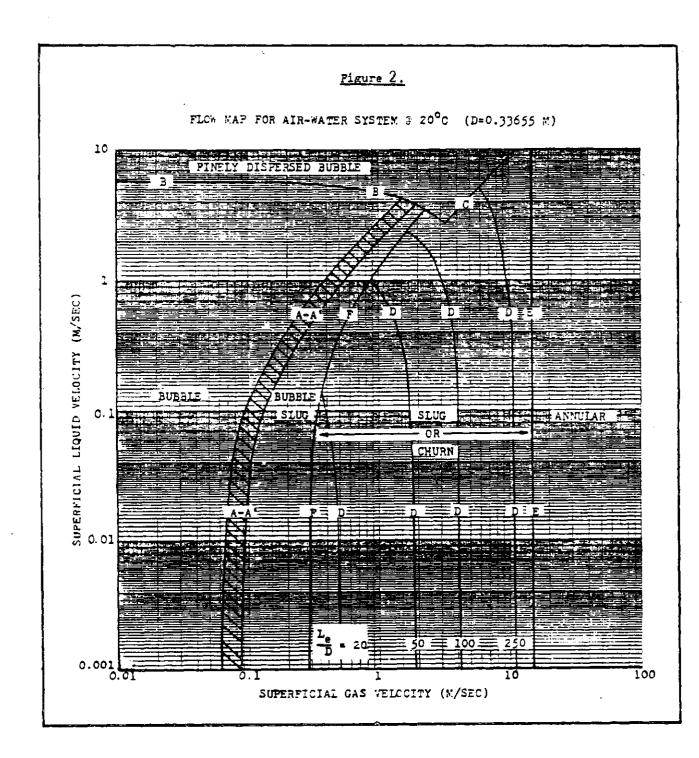


Figure 3 is a plot of apparent viscosity vs. superficial gas velocity in a 6 inch diameter column for air-aqueous CMC solution. For a given CMC concentration, ie; apparent viscosity, we can estimate the flow pattern of operation, as the superficial gas velocity is increased. This figure is useful for different apparent viscosities ranging from 0.01 to 0.12 Pa-sec.

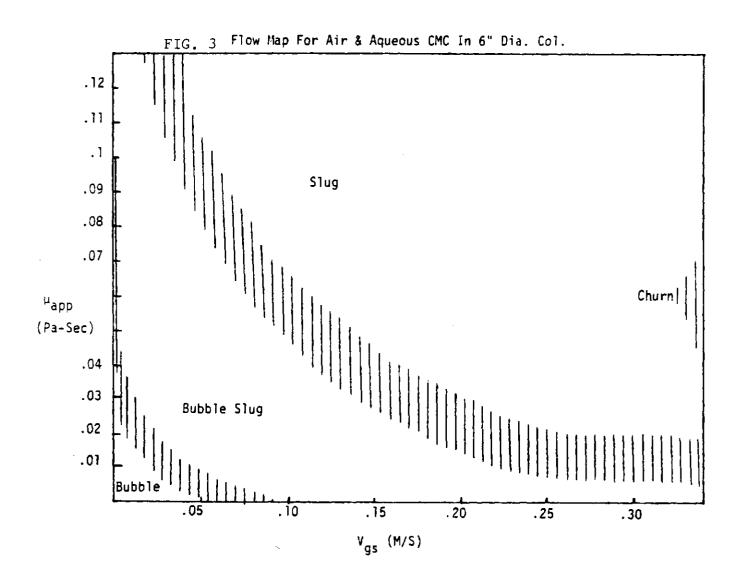


Figure 4 is a plot of weight percent CMC vs. superficial gas velocity for a 6 inch diameter column for air-aqueous CMC solution. Knowing the CMC concentration in weight percent, we can estimate the flow pattern of operation at different superficial gas velocities for a given CMC concentration. This figure is applicable for different CMC concentrations ranging from 0.25 to 2.0 weight percent.

Fig. 4 Flow Map For Air & Aqueous CMC In 6" Dia. Column

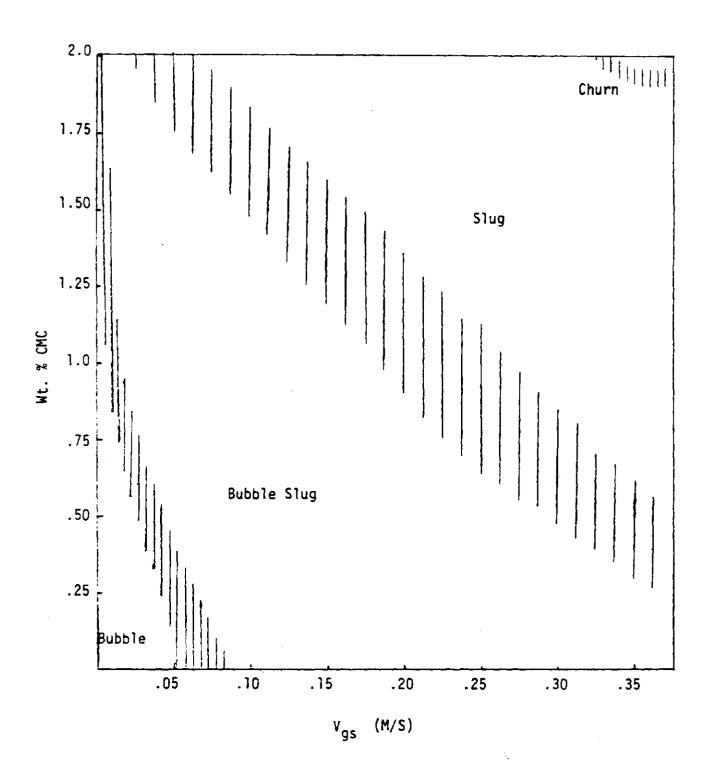


Figure 5 is a plot of superficial gas velocity vs. column diameter for air-water system. It gives the dependency of the flow pattern on superficial gas velocity and column diameter for air-water system. Knowing the superficial gas velocity and column diameter we can estiate the flow pattern of operation. This flow map is applicable to both small and large diameter columns ranging from 0.01 to 1m diameter.

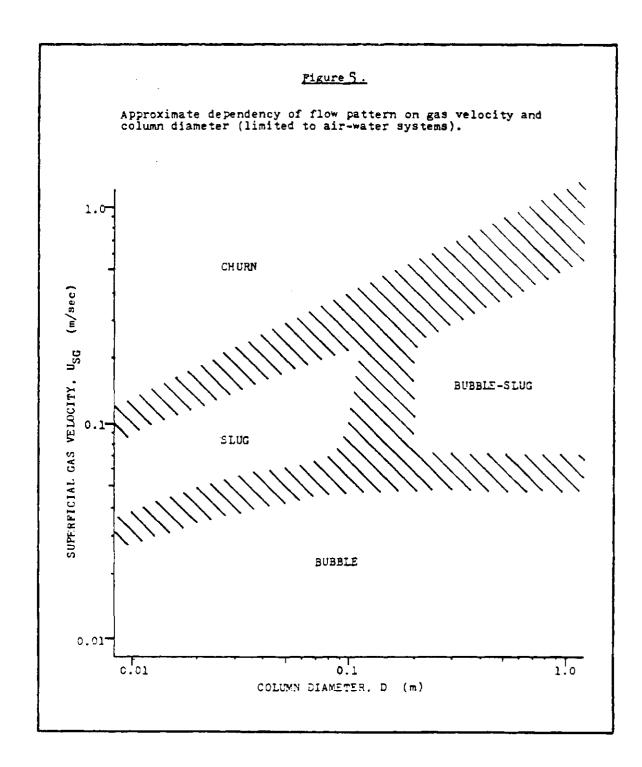


Figure 6 is a flow map proposed for coal liquids-hydrogen system at 450°C and 130atm in a 0.3048 m diameter column. Knowing the superficial gas and liquid velocities for the coal liquids-Hydrogen system, we will be able to estimate the flow pattern and its transition boundary. This also gives the slug or churn transition region for different length to diameter ratios. The block marked gives the flow pattern and the transition that bubble column reactors are expected to operate.

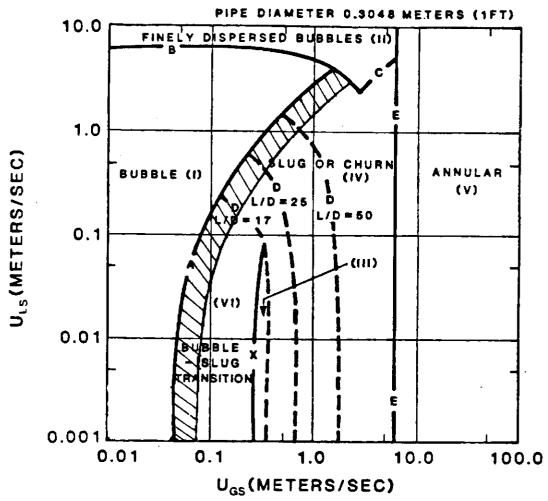


Figure 6 . Proposed Flow Map For Coal Liquids-Hydrogen, 450°C, 130 atms, 0.3048 m Diameter

Figure 7 is a flow map proposed for coal liquids-hydrogen system at  $450^{\circ}\text{C}$  and 130 atm in a 3.048m diameter column. This figure is similar to figure 6 but is applicable to a larger diameter column, ie; 3.048 meter diameter. Knowing the superficial gas and liquid velocities for the coal liquids-Hydrogen system, we will be able to estimate the flow pattern and its transition boundary. This also gives the slug or churn transition region for different length to diameter ratios. The block marked gives the flow pattern and the transition that bubble column reactors are expected to operate.

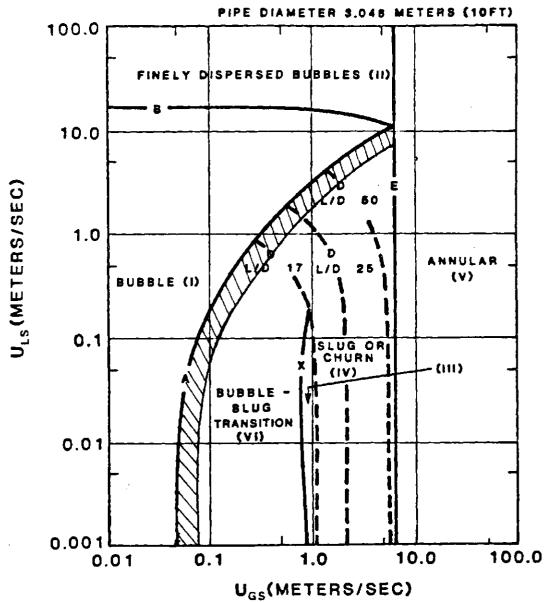


Figure 7. Proposed Flow Map For Coal Liquids-Hydrogen, 450°C, 130 atms, 3.048 m Diameter

#### NOMENCLATURE

 $d_B$  = Bubble size, cm

d<sub>Bl</sub> = Small bubble size, cm

 $d_{B2}$  = Large bubble size, cm

 $E_G$  = Gas holdup

 $E_{G1}$  = Holdup due to small bubbles

 $E_{G2}$  = Holdup due to large bubbles

VGS = Superficial gas velocity, M/S

Thic = Thickness of the gas distributor, Meters

Pore = Pore size of the gas distributor, Microns

a = Interfacial area, Cm<sup>-1</sup>

CMC = Carboxy Methyl Cellulose

IPA = Iso Propyl Alcohol

LLS = Length of liquid slug, meters

LTB = Length of Taylor bubble, meters

K = Power law parameter, consistency index, Pa-sec

n = Power law parameter, flow index

g = Acceleration of gravity

 $R_{o}$  = Column radius, meters

R<sub>f</sub> = Film radius, meters

 $R_{o}-R_{f}$  = Film thickness, meters

ν<sub>1</sub> = Velocity of small bubbles

V<sub>2</sub> = Velocity of large bubbles

 $V_{SB}$  = Volume of small bubbles

 $V_{LB}$  = Volume of large bubbles

 $V_T$  = Total volume

## GREEK LETTERS

e = Liquid density, gms/cc

 $\mu_{app}$  = Apparent viscosity, Pa-Sec

 $\dot{\gamma}$  = Shear rate, Sec<sup>-1</sup>