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MASTER

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Abstract

In modeling three-phase flow reactors, mass transfer, heat transfer, interfacial area, and dispersion of phases as well as hydrodynamics have to be investigated. Flow regions based on two-phase flow (gas-liquid) of bubble columns have been studied. However, most of the studies have been based on the air-water system, which may differ significantly from the coal liquid-H₂ system. The applicability of these studies will be tested using experimental data from coal liquefaction studies.

For three-phase fluidized bed reactors, flow regimes have not been well established. The published data and correlations will be used to examine their applicability to coal liquefaction processes.

OBJECTIVES

The overall objective of the proposed project is the establishment of mathematical models for the simulation, optimization, and evaluation of the various existing or planned liquefaction processes, for the determination of the relative merits of these processes, and for the identification of the areas in which further research is needed to make coal liquefaction a viable means to meet the nation's imperative energy need. In order to compare the various processes on a meaningful basis, the optimization and evaluation will be done in the context of a complete and self-contained liquefaction process. Peripheral units such as gasifier, combustor, shift-converter, separator, etc., will therefore be analyzed and simulated as well.

STATEMENT OF WORK

As the models for process units are the most lacking elements in the proposed project, a substantial portion of the two years' research effort will be devoted to the establishment of unit models. Special attention will be paid to the liquefier as it is the least understood part of the liquefaction process. A schedule for the following tasks is given at the end of this section.

Task I

- a. Review literature on the hydrodynamics of two-phase flow, and incorporate relevant material to the development of a liquefier unit model.
- b. Analyze the effect of coal minerals and other catalysts on the kinetics and mechanism of coal dissolution and hydrogenation.

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- c. Develop a kinetic and hydrodynamic model of coal dissolution and hydrogenation, and compare the model's predictions with experimental data.
 - d. Study the effect of the various operating parameters such as temperature, pressure, reactor size, and degree of mixing on the performance of the liquefier.
 - e. Study the transient behavior of the liquefier due to operational changes.

Task II

- a. Integrate unit models developed in Task I into process models for the simulation of selected direct coal liquefaction processes.

THREE-PHASE FLUIDIZATION

In the last period, the phase holdups, bubble behavior, and dispersion of phases were reviewed. Data and correlations were collected to examine the applicability of correlations. In this period, a survey of flow regime map, mass transfer, and heat transfer has been completed. This information should lead to a better understanding of design for a three-phase fluidized bed.

Flow Region Map

No complete flow regime map has yet been devised. Muroyama et al. (1978) studied three-phase fluidization with glass beads of six different sizes (0.215 ~ 6.9 mm), alumina beads (2.0 mm), and Raschig rings (5.2 mm) in columns of 6 cm and 10 cm in diameter. Flow patterns were classified

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into three regions: coalesced bubble flow ($U_{10} < 8$ cm/sec, $U_{g_0} < 10$ cm/sec); dispersed bubble flow ($U_{10} > 8$ cm/sec, $U_{g_0} < 10$ cm/sec); and slug flow ($U_{g_0} > 10$ cm/sec). These criteria qualitatively agree with Michelsen and Østergaard's data (1970). Additional effort is necessary to construct a more general flow regime map on a theoretical base.

In different flow regimes, phase holdups, phase mixings, mass transfer, and heat transfer will be different. A well defined flow regime map is required for choosing best correlations of parameters in each flow regime.

Mass Transfer

Gas-Liquid Interface Mass Transfer. The gas-liquid mass transfer depends upon the gas-liquid interfacial area, the gas residence time, and the gas-liquid mass transfer coefficient. The interfacial area plays an important but complex role on the mass transfer rate in the three-phase fluidized-bed system. Østergaard and Suchozebrski (1971) and Østergaard and Fósþól (1972) studied the relations between the bubble dynamics and volumetric mass-transfer coefficient ($k_L a_L$). Østergaard and Suchozebrski (1971) showed that $k_L a_L$ increased with increasing gas flow rate, but was not influenced by liquid velocity. Particle size was observed to affect the mass transfer rate tremendously. The volumetric mass transfer coefficients in a fluidized bed with 6-mm particles were about an order of magnitude larger than those obtained with 1-mm particles. The coefficients in the system with no particles present were between the above two cases. Østergaard and Fósþól (1972) showed that an increase in the superficial liquid velocity had no effect on the adsorption rate in beds of 6-mm particles and in solid-free

bubble columns. But in beds of 1-mm particles, an increase in liquid velocity caused a marked increase on the volumetric mass-transfer coefficients. Østergaard and Føsbøl (1972) also showed that the volumetric coefficient was a strong function of bed position. In solid-free bubble columns and in beds of 1-mm particles, the volumetric mass transfer coefficient decreased with increase in the distance from the gas distributor. In beds of 6-mm particles, the volumetric mass transfer coefficient first increased and then decreased with the distance from the gas distributor.

Table I lists all the systems used for studying gas-liquid mass transfer. The available correlations are listed in Table II.

Heat Transfer

The wall to bed heat-transfer coefficient in a three-phase fluidized bed was measured by Østergaard (1964), Viswanathan et al. (1968), Armstrong et al. (1976), Baker et al. (1978), and Kato et al. (1981). Armstrong et al. studied air-water fluidized bed with four different particle sizes (0.5 mm, 1 mm, 3 mm, and 5 mm). The superficial gas velocity ranged from 0 to 23.77 cm/sec and the superficial liquid velocities from 0.82 to 12.6 cm/sec. The results showed that heat transfer coefficient increased with superficial gas velocity for all liquid flow rates and particle sizes. For a given particle size, the heat transfer coefficient reached a maximum with respect to the bed porosity ($\epsilon_l + \epsilon_g$). This is similar to the behavior of the two-phase (liquid-solid) fluidized bed. In general, the maxima in the curves shifted to lower porosities with increasing particle size. Kato et al. (1981) suggested a correlation for wall-bed heat transfer coefficient:

$$Nu' = 0.044 (Re'Pr)^{0.78} + 2.0 Fr_g^{0.17}$$

where

$$Nu' = hd_p \epsilon_l / [k_l (1 - \epsilon_l)]$$

$$Re' Pr (= Pe') = d_p \bar{U}_l \rho_l C_{p_l} / [k_l (1 - \epsilon_l)]$$

$$Fr_g = \bar{U}_g^2 / (gd_p)$$

- h: heat transfer coefficient
- d_p : particle diameter
- ϵ_l : liquid holdup
- k_l : thermal conductivity of liquid
- \bar{U}_l : superficial liquid velocity
- \bar{U}_g : superficial gas velocity
- ρ_l : density of liquid
- C_{p_l} : specific heat of liquid
- g: acceleration of gravity

Table I

SUMMARY OF SYSTEMS STUDIED FOR GAS-LIQUID INTERFACE MASS TRANSFER

Investigators	System	Particle Size	Experiment Unit
Massimilla et al. (1959)	absorption of CO ₂ in water	silica-sand particles (0.22 mm) ballotini (0.5 ~ 0.8 mm)	
Adlington Thompson (1965)	absorption of O ₂ in sodium sulfite solution	glass particles (0.3 ~ 3 mm)	
Østergaard Suchozebrski (1971)	absorption of CO ₂ in water	glass ballotini (1 mm and 6 mm)	4 in ID. Height 2.91 m
Østergaard Pøshøj (1972)	absorption of O ₂ in water	glass ballotini (1 mm and 6 mm)	4 in ID. Height 2.91 m
Kito et al. (1976)	absorption of CO ₂ in aqueous sodium hydroxide solution	spherical beads (1.1, 2.65, 2.87 cm) ρ_p (0.89 g/m ³ , 1 g/m ³)	10 in ID. Height 150 cm
Nishikawa et al. (1977)	absorption of O ₂ into copper-catalyzed sodium sulfite solution	spherical particles (1.01, 2.59, 4.87 mm)	
Østergaard (1978)	absorption of CO ₂ in a carbonate/bicarbonate buffer	glass ballotini (1.1, 3.0, and 6.0 mm)	0.216 m ID. Height 8.50 m

Table II

EMPIRICAL CORRELATIONS FOR GAS-LIQUID INTERFACE MASS TRANSFER

Authors	Correlation
Strumillo Kudra (1977)	$A = 21.5 U_{og}^{0.92} L^{0.34} H_{st}^{0.83} d_w^{-0.94}, m^2/m^2$ <p> A = interfacial area per unit crosssection of the column (m^2/m^2) U_{og} = superficial gas velocity ($m s^{-1}$) H_{st} = static bed height (mm) L = liquid flow rate ($m^3 m^{-2} h^{-1}$) d_w = particle diameter (mm) </p>
Fukushima (1979)	$K_L^* a d_p^2 \epsilon / D_L \phi_g = 1.4 \times 10^4 Re_L^{0.38} Re_g^{-0.2} Sc_L^{1/2} (d_p/d_T)^{1/2}$ <p> $K_L^* a$ = volumetric mass transfer coefficient ϵ = total void fraction ($\epsilon_l + \epsilon_g$) D_L = diffusivity ϕ_g = gas holdup $Sc_L = \mu_L / \rho_L D$ d_T = column diameter </p>
Østergaard (1978)	$K_{2a} = 9.7 U_g h^{-1} \text{ bubble column} \quad 4.3 < U_g < 12.4, (\text{cm/sec})$ $K_{2a} = 4.7 U_g^{0.76} h^{-1} \text{ 1 mm particle} \quad 1.5 < U_g < 4.6$ $K_{2a} = 7.0 U_1 U_g^{0.25} h^{-1} \text{ 3mm particle} \quad 4.3 < U_g < 12.8$ $K_{2a} = 21.1 U_g^{0.93} h^{-1} \text{ 6 mm particle} \quad 4.7 < U_g < 12.7$ <p style="text-align: right;">$6.8 < U < 9.7$</p>

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BUBBLE COLUMN REACTOR MODELING

The bubble column, despite its simple construction, requires relatively complex reactor models. The residence time distribution of the liquid phase indicates intermediate mixing, which cannot be described by either plug flow or complete mixing models.¹ Therefore, the dispersion model or the cell model is recommended.²

The uncertainties in determining parameter values such as holdups, interfacial areas, and mass transfer coefficients add more difficulties in modeling the reactor for scale-up. Therefore, efforts are being made in reducing the uncertainties. An evaluation of prediction methods for the hydrodynamic parameter is in progress. Experimental data are collected and classified according to flow regimes. Calculated values from correlations are then compared with the experimental data in each flow regime. Appropriate correlations or theories can then be selected. A more detailed discussion on this type of analysis is given by Mandhane.³

There are other problems in modeling the bubble column. Deckwer¹ suggested that the reliable reactor model should take into account axial dependence of holdup and interfacial area as well as the variation of molar gas velocity. He also pointed out that correlations based upon measurements from nonreactive systems are unlikely to be reliable in industrially reactive systems. Considerations will be given to these areas in developing the reactor models.

Heat Transfer

Recently, Michael and Reichert⁴ measured the heat transfer coefficient in a bubble column with some liquid hydrocarbons and various polyethylene,

hydrogen dispersions with solid contents up to 54 weight %. They found increasing the solid concentrations does not always increase heat transfer. This was due to the large increase in viscosity which accompanied the increase in solid concentration. Heat transfer was found to decrease with increasing viscosity by Kolbel.⁵

The equation derived theoretically by Deckwer⁶ using the surface renewal model of mass transfer and Kolmogoroff's theory of isotropic turbulence was found satisfactory in fitting the data. The superficial gas velocity where data were obtained varied from 0.9 - 11.8 cm/s which was approximately the range of applicability suggested by Deckwer.

The N_2 /paraffin/ Al_2O_3 system was also investigated by Deckwer.⁷ Using the equation he developed, he was able to fit the data. Based upon these two investigations of slurry systems, the equation formulated by Deckwer seems most appropriate.

It is important to note that the estimation of physical properties gives errors that are comparable to the deviations of calculated value from experimental values. Therefore, the prediction of physical properties plays an important role in furthering the accuracy of heat transfer correlation in slurry systems.

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FUTURE WORK PLAN

The literature survey on hydrodynamics and mass and heat transfer parameters will be periodically updated. The literature search on the multi-phase flow reactor model and on the kinetic and physical properties of coal systems will be continued. Evaluation of parameter prediction methods will be completed, and suitable reactor models will then be identified.