

HYDRODYNAMICS, MIXING AND MASS TRANSFER IN VARIOUS SIZED REACTORS

Introduction:

The performance of a coal liquefaction reactor depends not only on the intrinsic kinetics but also on the prevailing hydrodynamic, mixing, and mass transfer characteristics of the reactor. The scaleup of the reactor will depend on the reproducibility and predictability of these characteristics in various sized reactors. The performance of a coal liquefaction reactor depends upon parameters such as gas and slurry holdup, the distribution of the gas in the reactor, axial and radial mixing (both mass as well as heat) within the reactor, the importance of mass transfer resistance at the gas-liquid interface, both radial and axial distributions of the catalyst (if used), and finally, on the intraparticle diffusion effect from the catalyst, if any. A proper model of a coal liquefaction reactor requires that these characteristics be expressed as a function of reactor scaleup variables such as gas and slurry velocity, reactor diameter, length to diameter ratio of the reactor, nature of the gas distributor, and the nature of any other internal or external circulation devices used in the reactor. The available literature for simple air-water two and three phase bubble columns for these hydrodynamic, mixing, and mass transfer characteristics is quite extensive. The data obtained from documents in the LTDB (Liquefaction Technology Data Base) and open literature will be used for evaluating the applicability of the concepts to the design, modeling, and scaleup of coal liquefaction reactors.

Coal Liquefaction Reactors

A typical coal liquefaction unit consists of a preheater and a reactor. The transfer-line slurry preheater raises the temperature of the coal-oil slurry to the desired reactor inlet temperature and simultaneously causes thermal degradation and dissolution of a large fraction of coal to occur. The preheater is usually a radiant fired helical coil heater, wherein the coal-oil slurry mixed with hydrogen is introduced. Reciprocating plunger pumps have been used to pressurize the feed slurries to the preheater (Gorrin, 1982). In the reactor itself, coal dissolution is completed, and hydroprocesses such as hydrogenation, hydrocracking, hydrodesulfurization, hydrodenitrogenation, etc. take place. Since the thermal degradation of the coal in slurry begins in the preheater, it can be regarded as an extension of the reactor. Although extensive work has been done in order to obtain design data for large scale liquefaction-process preheaters (Gorrin, 1982; Baumert and Bornett, 1982; Traeger et al., 1979; U.S. DOE, 1979; Shah et al., 1978), it is very difficult to interpret them because the flow regimes in the preheater are not well understood, nor are the physical property changes fully defined. For the thermal hydrodynamic modeling, the preheater can be divided into three regions. The temperature increases as the slurry passes through the preheater. In region I, the viscosity passes through a minimum value. Region II is characterized by a sharp increase in viscosity due to swelling and agglomeration. The region ends when significant chemical reaction and dissolution occur. In region III, depolymerization and dissolution of the gel take place, and the viscosity rapidly decreases. The flow regime in the first region is turbulent-laminar or laminar-laminar, in the second region it is laminar, while in the third region it is turbulent two phase non-Newtonian flow.

temperature. In this report, we will only consider the slurry reactor models for isothermal reactors and the scaleup aspects associated with isothermal reactors.

Though easy to use, bubble column reactors are difficult to design because of the complexity of flow characteristics and their unknown hydrodynamic behavior under different sets of design parameters. The proper design and scale-up of a bubble column slurry reactor requires a good mathematical model. Ledakowicz et al. (1983) have outlined a design procedure. The application of the procedure requires an exact definition of the requirements, i.e. the required production level, nature of the reaction system, and the type of product yield structure. These quantities permit a first choice of the adjustable operational conditions which include phase velocities, temperature, pressure, and direction of the flows. In addition, data on physical properties of the reaction mixture and its components and phase equilibrium data as well as kinetic parameters are needed. Based on these data, non-adjustable parameters are calculated using the available literature correlations. Knowing these parameters, the mass, momentum and heat balance equations are solved to calculate product yield, productivity, concentration and temperature profiles. If possible, experimental verification is provided by collecting the relevant data on the pilot plant.

For any multiphase reactor design, non-adjustable design parameters constitute the important group of parameters which depend on the chosen reactor geometry and the adjustable operating conditions as well as the process data. These interrelated non-adjustable parameters include phase holdups, the interfacial areas, the heat and mass transfer properties, and the dispersion coefficients. The reactor geometry, reaction parameters, and the adjustable and non-adjustable parameters are then introduced in the

fundamental reactor model equations derived on the basis of the physical and chemical phenomena occurring in the reactor.

Several problems encountered in the design of the coal liquefaction reactor are as follows:

- a. Estimation of the process specific data, particularly, solubilities and kinetic quantities.
- b. Accessibility of the non-adjustable operating conditions which must be estimated from empirical correlations established from cold flow studies and not from conditions prevailing in the coal liquefaction process.
- c. Choice of a correct reactor model should be based on two factors: realistic judgment of possible improvements due to the usage of complex models and the uncertainties in process data.

The major physico-chemical phenomena important for modeling the bubble column slurry reactor are:

- a. Gas-liquid mass transfer
- b. Liquid-solid mass transfer
- c. Reaction rate
- d. Heat transfer
- e. Heat effects such as hot spots and runaway possibilities
- f. Particle settling
- g. Mass dispersion in gas phase
- h. Mass and heat dispersion in liquid phase
- i. Vaporization
- j. Variation of gas throughput accompanied by changes in phase holdups, interfacial area, etc.

Few papers concerning the fundamentals of coal liquefaction reactor modeling have been published (Lee et al., 1978; Shah et al., 1978; Han et al.,

1978; Parulekar and Shah, 1980; Shah and Parulekar, 1982; Nunez et al., 1982). The proposed models consider most of the above phenomena. The number of differential equations representing the model depends on the type of kinetic model assumed. Even for simple kinetic expressions, the number of equations is large, and the computation efforts are considerable. For example, the model developed by Lee et al. (1978) consists of 6 first order differential equations for the preheater and 6 second order differential equations for the dissolver. Only a mass balance was considered for the development of these equations. Parulekar and Shah (1980) also proposed a multiphase reactor model of a complex nature.

Reactor Models:

Several lumped kinetic models for donor-solvent coal liquefaction are reviewed in the next section. The complexity of the kinetic model depends upon the number of lumped species used and the chosen reaction paths. Our knowledge in the area of kinetic modeling is in the development stage, and hence a simple lumped kinetic model featuring basic reaction paths will be used in describing the reactor model.

The simple reaction path network which is most commonly used to describe the kinetics is as follows (Cronauer et al., 1978):

If A denotes the heavy distillate (which contains asphaltenes, preasphaltenes and the reactive organic matter in the coal), B denotes the light distillate and C denotes the volatile products; the reaction mechanism can be described as shown below:

Reaction 1:



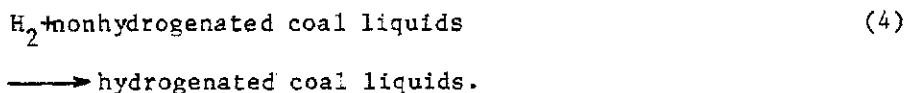
Reaction 2:



Reaction 3:



Reaction 4:



Here ℓ and g denote liquid and gas phases, respectively. In general, all reactions are partly thermal and partly catalytic. Species A and B are the mixtures of hydrogenated and nonhydrogenated coal liquids.

Axial-Dispersion Model for an Isothermal Reactor:

Many highly sophisticated models are available in literature. The model equations, based on the first principles, are usually solved numerically to analyze the complex scale up problems. A typical example of such a complex model will be outlined in this section.

Parulekar and Shah (1980) described such a model for an isothermal reactor. The model assumed:

- (a) The gas phase is an ideal gas.
- (b) The reactor is isobaric (since the total pressure drop is not significant as compared to the total pressure).
- (c) The axial dispersion model is applicable to nonvolatile liquid and solid components.
- (d) For simplicity, all reaction rates are directly proportional to the catalyst concentration.
- (e) The gas phase moves in plug flow.
- (f) The reactor is the cocurrent upflow type.
- (g) All the global reaction rates can be expressed in terms of the gas phase concentration of hydrogen. This means that the gas-liquid interface mass-transfer resistance is negligible. r_1 represents rate of the reaction

(1) in $\text{kg/m}^3 \text{ liq. sec.}$, and r_2, r_3, r_4 represent the rates of the reactions

(2), (3), and (4) in $\text{kg-mol/m}^3 \text{ liq. sec.}$, so that

$$r_1 = k_1^* e^{-E_1/R_g T} C_{Al} C_{H_2} C_s, \quad i = 1, 2 \quad (5)$$

$$r_3 = k_3^* e^{-E_3/R_g T} C_{Bl} C_{H_2} C_s, \quad (6)$$

$$r_4 = k_4^* e^{-E_4/R_g T} C_{H_2} C_s \quad (7)$$

(8) There are no radial variations in phase holdup and the concentration of the reacting species.

Parulekar and Shah (1980) have established the following model equations:

$$z = x/L,$$

$$\bar{U}_g = \frac{U_{og}}{(U_{og})_o}, \quad \bar{U}_l = \frac{U_{ol}}{(U_{ol})_o}$$

$$H' = C_{H_2}/C_T, \quad C_T = P/R_g T, \quad C = 1 - H'$$

$$A = C_{Al}/\rho_l, \quad B = C_{Bl}/\rho_l,$$

$$Pe_l = \frac{(U_{ol})_o L}{(E_{zl})_o}, \quad Pe_s = \frac{(U_{ol})_o L}{(E_{zs})_o} \quad (8)$$

$$R_1 = \frac{k_1 \rho_l L}{(U_{ol})_o}, \quad R_2 = \frac{k_2 \rho_l L}{(U_{ol})_o}, \quad R_3 = \frac{k_3 \rho_l L}{(U_{ol})_o}, \quad R_4 = \frac{k_4 L}{(U_{ol})_o}$$

$$r = \frac{(U_{ol})_o}{(U_{og})_o}$$

$$f = \frac{U_t}{(U_{ol})_o}, \quad f_1 = \frac{U_t'}{(U_l)_o}$$

$$e_s = \frac{E_{zs}}{(E_{zs})_o}, \quad e_l = \frac{E_{zl}}{(E_{zl})_o}$$

Here $k_i = k_i^* e^{-E_i/R_g T}$ for $i = 1, 2, 3, 4$.

The relevant equations for this model could be represented by the following set of equations.

Gas-phase hydrogen

$$\frac{d(U_g H')}{dz} = -R_4 r H' C_s e_l \quad (9)$$

Gas velocity

$$\frac{dU_g}{dz} = (R_2 A + R_3 B - R_4) r H' C_s e_l \quad (10)$$

Liquid phase A.

$$\frac{1}{Pe_l} \frac{d}{dz} \left(e_l \frac{dA}{dz} \right) - \frac{d(U_l A)}{dz} - R_1 A C_s e_l \quad (11)$$

$$- \frac{R_2 A H' C_s M_l C_T}{\rho_l} = 0$$

Liquid phase B.

$$\frac{1}{Pe_l} \frac{d}{dz} \left(\bar{e}_l \frac{dB}{dz} \right) - \frac{d(\bar{U}_l B)}{dz} + R_1 A C_s \epsilon_l \quad (12)$$

$$\frac{-R_3 B H' C_s \epsilon_l M_C C_T}{\rho_l} = 0$$

Liquid Velocity

$$\frac{d\bar{U}_l}{dz} = \frac{C_T}{\rho_l} (R_4 M_{H_2} - (R_2 A + R_3 B) M_C) H' C_s \epsilon_l \quad (13)$$

Solid balance

$$\frac{1}{Pe_s} \frac{d}{dz} \left(\bar{e}_s \frac{dC}{dz} \right) - \frac{d}{dz} [(\bar{U}_{sl} - \bar{U}_t) \epsilon_{sl} C_s] = 0, \quad (14)$$

where

$$\bar{U}_{sl} = \bar{U}_l \left(\frac{\rho_l}{\rho_{sl}} \right) - \frac{\rho_s \epsilon_s}{\rho_{sl}} f.$$

Depending upon the relative magnitudes of absorption of hydrogen and production of volatile products, (C), the gas velocity may either increase or decrease. Any change in the gas velocity is obtained by considering a total gas balance over a differential section of the reactor. Equation 10 results from the total gas balance. The principle of the conservation of mass implies that the total mass of the three phase mixture is the same everywhere in the direction of the flow. Since it is assumed that almost all the chemical changes occurring in the reactor are in the liquid phase, it is obvious that the mass of the gas-liquid mixture should remain axially constant. The change

in liquid velocity is thus related to the change in the gas velocity and subsequently to the rates of individual reactions. The equation for the change in liquid velocity is given by equation 13.

The initial conditions for equations 9, 10, and 13 are

$$H' = H_0', \quad \bar{U}_g = 1, \quad \bar{U}_l = 1 \text{ at } Z = 0^+ \quad (15)$$

The boundary conditions for Equations 11, 12, 14 are at $Z=0^+$:

$$\frac{dA}{dZ} = Pe_g (A - A_0) \quad (16a)$$

$$\frac{dB}{dz} = Pe_g (B - B_0) \quad (16b)$$

$$\frac{dC_s}{dz} = \frac{Pe_s}{e_s} [(U_{si} - f_l)(1 - \epsilon_g)(C_s - C_{so})] \quad (16c)$$

at $Z=1^-$

$$\frac{dA}{dz} = \frac{dB}{dz} = \frac{dC_s}{dz} = 0 \quad (17a)$$

$$C_{si} = C_{so} \quad (17b)$$

The boundary condition (17b) is obtained from an overall solid-phase mass balance. C_{so} , the catalyst concentration in the feed, is given by

$$C_{so} = \frac{W_s \rho_l}{1 - \frac{\rho_s - \rho_l}{\rho_s} W_s} \quad (18)$$

Overall yields and consumptions of various components can be based on the amount of slurry entering the reactor (expressed as amount produced or consumed per unit of slurry fed). The yields and consumptions can be expressed in terms of dimensional and dimensionless variables as follows:

Consumptions

$$Y_A = A_0 - (A \bar{U}_L)_1, \quad (19)$$

$$Y_{H_2} = [H_0 - (H \bar{U}_g)_1] C_{T H_2} / r_{p_2}, \quad (20)$$

where Y_A and Y_{H_2} are the consumptions of A and H_2 , respectively.

Yields.

$$Y_B = (B \bar{U}_L)_1 - B_0 \quad (21)$$

$$Y_C = [(C \bar{U}_g)_1 - C_0] C_T M_C / r_{p_2} \quad (22)$$

where Y_B and Y_C are the yields of B and C, respectively. Subscript 1 denotes the values of variables at the reactor outlet.

To solve the above set of model equations, all the adjustable and non-adjustable design parameters should be known. The dependency of these design variables can be obtained through empirical or theoretical correlations suggested for bubble column slurry reactors.

The section on the results pertaining to the bubble column slurry reactor modeling will be presented after an extensive review of the required estimation of the non-adjustable design parameters.

Design Parameters Estimations for Bubble Column Slurry Reactors:

Flow regime, bubble size distribution and coalescence characteristics, phase holdup, interfacial mass transfer coefficients, gas-liquid interfacial area, dispersion coefficients, heat transfer characteristics, and the concentration profile of solids are important design variables for bubble column reactors. A thorough knowledge of these interdependent parameters is necessary for the proper scaleup of these reactors.

a) Flow Regimes:

The hydrodynamic, transport, and mixing properties strongly depend on the prevailing flow regime. Many investigators (Govier and Aziz, 1972; Lockett and Kirkpatrick, 1975; Wallis, 1969; Kawagoe et al. 1976; Hills, 1976; Miller, 1980) have proposed different criteria to differentiate flow regimes. Wallis (1969) has characterized the upward movement of the bubble swarms into three separate flow regimes.

Bubbly flow: This regime is characterized by almost uniformly sized bubbles with equal radial distribution.

Churn turbulent flow: At higher gas velocities the homogeneous gas-liquid dispersion cannot be maintained and an unsteady flow pattern with channeling occurs.

Slug flow: In small diameter columns at high gas flow rates, large bubbles are stabilized by the column wall leading to the formation of bubble slugs.

Following Kurten's suggestion (1982), the typical dimensions of an industrial scale coal liquefaction reactor are given as: diameter $d_R > 3m$, length $L > 12m$, and superficial gas and liquid velocities; $U_G = 0.02-0.1$ m/s and $U_L = 0.002-0.005$ m/s, respectively. For this industrial size reactor, the churn turbulent regime might set in at high gas velocities. Knowledge of the

transition from bubbly flow to churn turbulent flow is very important because the achievable conversion strongly depends on the flow regime. Schumpe et al. (1979) have recommended that churn turbulent flow be avoided. Kara et al. (1982) studied the transition in flow regime for coal slurries over a wide range of gas and liquid velocities. For the gas and liquid velocities under consideration for aqueous coal slurries, a transition flow regime (bubbly-churn turbulent) will be expected. In the SRC-II coal liquefaction medium, a surface tension ≤ 10 dyne/cm is expected. Furthermore, the coal liquefaction reaction medium may behave as a mixture of polar organic compounds rather than as pure organic liquids. Shah et al. (1982a, 1982b) have studied the hydrodynamic and mixing characteristics of dilute alcohol solutions in two different sized bubble column reactors. Based on these studies, it seems reasonable to assume that for coal liquefaction reaction medium bubbly flow regime is possible for the gas velocity under consideration. There are no flow regime data available in the high temperature, high pressure pilot scale reactors.

b) Bubble dynamics: Bubble size, bubble rise velocity, bubble size distribution, and liquid and bubble velocity profiles have a direct bearing on the performance of bubble columns. For estimating the bubble diameters in bubble columns, the correlation of Calderbank (1967) is recommended. The bubble rise velocity can be estimated by the method proposed by Clift et al. (1978). This basic information helps in the flow regime characterization.

c) Phase holdups: Phase holdups are some of the most important parameters characterizing the hydrodynamics of bubble columns. The presence of solids does not affect the gas holdup significantly if the particles are well suspended. Recently Ying et al. (1980) applied the correlation of Akita and Yoshida (1973) to their data and concluded that the correlation of Akita

and Yoshida is equally adequate for three phase systems. Phase holdups (gas, liquid, and solid) might vary with respect to the radius and length of the reactor. If the solids are well suspended, then the slurry could be treated as a homogeneous phase and the gas holdup will be the only variable. The uneven distribution of gas holdup and/or slurry holdup can create significant problems due to the hot spots. Hills (1974) and Ueyama et al. (1980) have reported pronounced radial holdup profiles. Kobayashi et al. (1970) proposed an empirical correlation to evaluate the radial distribution of gas holdups. Insufficient data are available to evaluate the radial holdup profiles accurately for the three phase systems. In addition, axial variation in the holdups is possible if the amount of reacting gas absorbed is unequal to the amount of vapors or gaseous products formed. The axial variation in holdups is usually incorporated in the bubble column reactor model. Due to insufficient data and unavailability of accurate predictions, the radial variation in phase holdups is usually neglected.

As mentioned earlier, the coal liquefaction reaction medium has low surface tension and might have properties similar to polar organic mixtures. Shah et al. (1982a,b) have studied dilute alcohol solutions only up to C_4 alcohols and they report very high gas holdups. According to our best judgment, the dilute alcohol solutions which are surfactant solutions will give much higher phase holdups than the coal liquefaction reaction medium. The correlations of Akita and Yoshida (1973) and Bach and Pihofler (1978) are recommended for conservative estimations of gas holdup in direct coal liquefaction media.

Exxon Research and Engineering Company has reported some studies (1981 a,b) of gas holdup at high pressures. Their gas holdup data revealed that gas holdups strongly increased with pressure (<2000 psi). In addition, the gas

holdups showed a maximum with respect to superficial gas velocity in a 1 ton/day and a 250 ton/day pilot plants. It was observed that maximum gas holdup in the 250 ton/day reactor occurred at lower superficial gas velocity and had a higher magnitude. The data in the 250 ton/day reactor were taken at a temperature of 180°C. Further, the gas holdup data in the 1 ton/day reactor indicated no effect of liquid velocity on the gas holdup up to superficial liquid velocities of 0.6 cm/s. The reported gas holdup values are much higher than the ones predicted by the correlations of Akita and Yoshida (1973) and Bach and Pilhofer (1978). These gas holdups at higher pressure and temperature indicate a need to make systematic measurements of gas holdups at high pressures and temperatures in the pilot plant scale reactors.

d) Volumetric gas-liquid mass transfer coefficient: For determination of the volumetric gas-liquid mass transfer coefficients in two phase systems, Shah et al. (1982) have recommended the correlation of Akita and Yoshida (1973).

Singh and Carr (1983) have presented a relation between the volumetric hydrogen mass transfer coefficient ($k_L a$) and specific power consumption in mixing (E/V_R) in SRC-II process slurry reactors. They claim that their correlation is independent of reactor configuration. They also compare their correlation with other literature correlations such as Deckwer (1980), Perry and Chilton (1973), and Joosten et al. (1977). It should be carefully noted that their correlation is based on only two data points measured for the hydrogen-tetralin system in an unconventional CSTR pressure vessel (Kara, 1981). We feel that their correlation will only estimate the volumetric mass transfer coefficient within an order of magnitude. The mixing energy concept is not valid if the similarity between the stirred cell and the bubble column slurry reactor is not maintained.

In the three phase bubble column reactors, $k_L a$ can be affected by the presence of solids. Investigations of various authors (Voyer and Miller, 1968; Slessor et al., 1968; Kato et al., 1973; Juvekar and Sharma, 1973; Sittig, 1977) indicate that the degree of influence of suspended particles on $k_L a$ depends on the particle concentration, the particle size, the liquid-solid difference, the geometrical sizes, and the operating conditions of the reactor. For the coal liquefaction reaction medium one can expect negligible effect of solids on $k_L a$ in comparison with their two phase values. In the coal liquefaction reactor, we do not expect mass transfer resistance; and, hence, these values are not actually used in the design calculations. They serve as a guideline to check the assumptions regarding the kinetic controlling regime.

e) Backmixing in the liquid phase:

The extent of backmixing of the liquid phase is an important consideration in the design and scale-up of SRC-II reactors. Wen and Han (1975), Shah (1979), Shah et al. (1978), Joshi and Shah (1980) extensively reviewed the literature, existing models, and methods of measurement in two- and three-phase systems.

A large number of macromixing models have been proposed to correlate the residence time distributions in multiphase systems. These models can be simple, single parameter models, such as the axial dispersion model, or they can be more complex two-, three-, or four-parameter models. So far, the axial dispersion model has been the most widely examined model; however, it has not been applied sufficiently to systems other than air-water. Based on the available data, parameters of complex flow models (circulation cell model, tank in series model, residence time distribution model, Rice et al. model,

1980) cannot be evaluated in sufficient detail to be useful for design or scaleup purposes.

The reported data on the liquid phase axial dispersion coefficient, D_L , indicate that D_L depends on the gas velocity and column diameter. The influence of liquid phase properties is not clearly understood. The effect of liquid velocity is insignificant within the range of industrial interest (<0.03 m/s) though at high liquid velocities, Kara et al. (1981) observed some effect of liquid velocity.

Shah et al. (1982b) have recommended the use of the correlations of Joshi (1980) and Deckwer et al. (1974) for estimation of the axial dispersion coefficient for the liquid phase. Field and Davidson (1980) measured the liquid phase dispersion in an industrial bubble column of 3.2 m diameter by 19 m height. Their results confirmed the applicability of the correlations of Deckwer et al. (1974) and Joshi (1980) for estimating D_L .

Recently, Parimi and Pitchford (1982) measured the liquid phase dispersion coefficients in a 0.3 m diameter, 7.62 m tall bubble column for air-water. They found substantially less backmixing present in the column than predicted by Deckwer et al.'s correlation (1974). Parimi and Pitchford (1982) analyzed their data using the method of moments: the second moment is related to liquid phase Peclet number which characterizes the degree of backmixing in the vessel. It seems that with the use of more refined techniques such as regression analysis or multiparameter residence time distribution models, accurate determination of liquid phase dispersion coefficient would have been possible. In spite of the questionable analysis of data put forth by Parimi and Pitchford (1982), there seems to be a justified concern regarding the use of the axial dispersion model for industrial scale reactors.

Panvelkar et al. (1982) have presented residence time distribution data for the liquid phase in a dissolver in a demonstration scale solvent refined coal (SRC) plant operating around 728 K and 13.8 MPa. The axial dispersion model is fitted to the detector outputs to evaluate the Peclet number. In their analysis, they obtained a dispersion coefficient lower than the one obtained by using the correlation of Deckwer et al. (1974) by a factor of 11.4. It should be noted that they had only one point, and the errors in slurry velocity and background noise will alter the results.

Shah et al. (1982a) have measured the liquid phase dispersion coefficients in dilute alcohol solutions. They have reported that it would be premature to conclude that the correlations such as Deckwer et al.'s (1974) are not valid at the conditions of coal liquefaction, but it does seem clear that there is a need to study backmixing at elevated temperatures and pressures and for systems with physical properties closer to those of industrial systems.

The effect of solid particles on the dispersion coefficient is not clearly understood due to the lack of enough experimental data in the region of industrial interest. Ying et al. (1980) observed large discrepancies between actual and theoretical values of dispersion coefficients predicted with the help of different existing correlations.

f) Gas Phase backmixing:

Mixing in the dispersed gas phase was measured by Diboun and Schugerl (1967), Carleton et al. (1967), Towell and Ackerman (1972), Pilhofer et al. (1978), Mangartz and Pilhofer (1980), and Field and Davidson (1980). Compared to liquid phase dispersion, this data revealed considerable scatter.

Based on their recent data, Pilhofer et al. (1978) and Mangartz and Pilhofer (1980) have concluded that the bubble rise velocity in the swarm is a

characteristic variable which is mainly responsible for the gas phase dispersion. Field and Davidson (1980), who conducted measurements on gas phase dispersion in a large diameter column (thirty-two times that used by Pilhofer et al. (1978)), slightly modified the original equation of Pilhofer et al. (1978). Although the predictions of correlations proposed by Mangartz and Pilhofer (1980) and Field and Davidson (1980) may differ by up to 50%, both correlations are recommended due to the lack of sufficient literature data. A knowledge of the gas phase backmixing is essential for the estimation of gas phase Peclet number used in the design equations.

g) Solid phase dispersion coefficient:

The behavior of solid particles in the bubble column is conveniently interpreted by the one-dimensional dispersion model. Imafuku et al. (1968) have reported uniform radial distribution of solid particles for batch as well as continuous operation. Imafuku et al. (1968) have also reported that the dispersion coefficient of suspended solid particles is the same as that of the liquid within the column without solid particles. By measuring the concentration profiles under cocurrent and batch conditions, Kato et al. (1972a, b) proposed a correlation for the solid dispersion coefficient. This correlation is similar to the one proposed by Kato et al. (1972b) for the liquid phase dispersion coefficient. Indeed, D_L and D_S differ only slightly. As a guideline it can be assumed that $D_L = D_S$. The correlation of Kato et al. (1972a, b) could be modified by introducing a particle Reynolds number. Shah et al. (1982) modified this correlation based on extensive measurements in three phase bubble columns of different diameters. Therefore, use of this correlation is recommended. Many bubble column reactor models consider the solids to be completely mixed in the liquid phase and consider only a homogeneous slurry phase.

Recommendations for Design Parameter Estimations:

The literature search regarding the heat transfer coefficients, thermal dispersion coefficient, heat of reaction, etc. are presented in a separate section discussing the adiabatic reactor models and the hydrogen consumption rates. All the recommended correlations for the estimation of non-adjustable design parameters are given in Table I-1. In the case where a unique correlation is not recommended, the correlations will cover the range of possible values. We feel that for the estimation of parameters for the SRC-II reactor, these correlations are the best available. We further recommend that some design parameters measurements in high pressure, high temperature large bubble column reactors be made in order to have a higher degree of confidence in the design parameter estimations.

Application of bubble column slurry reactor models:

In addition to the estimations of design parameters such as phase holdups, dispersion coefficients, volumetric mass transfer coefficients, etc., more information on solids distribution is needed to solve the set of model equations given under section 3.

a) Solid Distribution: Cova (1966) proposed a model to predict the catalyst concentration profile in the reactor. This model can only be applied at low catalyst loading, small particle size, and constant liquid- and gas-phase superficial velocities. When superficial gas or liquid velocities and the gas and liquid holdups change in the direction of flow, this model cannot be applied. Parulekar (1979) proposed a model which is applicable under hindered-settling conditions and can be applied over a large range of catalyst loading and particle sizes. In the case of a cocurrent upflow reactor, the flux, n_s , is given by

$$n_s = \frac{C_s (n_s + n_l)}{\rho_{sl}} - C_s U'_t - \frac{E_{zs}}{(1-\epsilon_g)} \frac{dC_s}{dx} \quad (23)$$

Here n_l is the mass flux of liquid. Both n_g and n_l are based on the cross-sectional area occupied by slurry.

The settling velocity of the particle with respect to the column is related to that with respect to liquid by the relation

$$U_t' = U_t \frac{\epsilon_l}{1-\epsilon_g} \quad (24)$$

The parameters liquid holdup, slurry density, ρ_{sl} , and slurry velocity, U_{sl} , are defined as

$$U_{sl} = \frac{n_s + n_l}{\rho_{sl}} = U_l \left(\frac{\rho_l}{\rho_{sl}} \right) - \frac{\rho_s \epsilon_s U_t}{\rho_{sl}} \quad (25)$$

$$\epsilon_l = (1-\epsilon_g) \left(1 - \frac{C}{\rho_s} \right) \quad (26)$$

$$\rho_{sl} = \frac{\rho_s \epsilon_s + \rho_l \epsilon_l}{1-\epsilon_g} \quad (27)$$

To show the dependency of the overall yield and conversions on the adjustable parameters, the design equations for bubble column slurry reactors are solved. The treatment of Parulekar (1979) and Parulekar and Shah (1980) will be used as an illustrative example. The values (Parulekar, 1979) of physical properties of the three phases and the kinetic parameters for the reactions are listed in Table I-2, and the operating conditions are listed in Table I-3. The values of the kinetic parameters (Table I-2) for the reaction (equation 4) were chosen so that the hydrogen consumption was around 3 to 5% of the heavy distillate. The values given in Tables I-2 and I-3 are representative of the basic features of SRC II reactors.

Parulekar (1979) studied the effect of various operating conditions and the reactor configuration on the yields of light oil (B) and volatile products (C), consumption of hydrogen and heavy oil (A), and effective total vaporization.

b. Effect of feed temperature: Parulekar (1979) found that for a reactor temperature of 573-673 K, the consumption of heavy oil (A) and hydrogen increased with temperature, whereas the yield, Y_B , passed through a maximum with respect to temperature. The magnitude of the average superficial liquid velocity decreased with respect to increasing temperature. The catalyst concentration within the reactor increased with temperature. Based on the data obtained from several pilot plants (Fort Lewis, Wilsonville, Gulf R&D), Jones (1979) concluded that hydrogen consumption linearly increases with the reaction temperature. This observation is in qualitative agreement with the hydrogen consumption-temperature plot of Parulekar (1979) and the experimental results of Shah et al. (1978).

c. Effect of pressure: An increase in operating pressure increases the hydrogen consumption and the formation of C due to an increase in hydrogen concentration. These predictions are in qualitative agreement with the experimental observations.

d. Effect of liquid velocity: An increase in liquid velocity decreases liquid phase residence time and increases Peclet numbers of both the liquid and solid phases and also reduces the importance of settling of the catalyst particles. With an increase in liquid velocity, the catalyst concentration gradient throughout the reactor decreases. Both Y_{H_2} and Y_C decrease, whereas Y_B goes through a maximum as the liquid velocity is increased.

e. Effect of superficial gas velocity: An increase in superficial gas velocity results in increased backmixing in the liquid and solid phases, which causes a decrease in the local catalyst concentration. Consumption of A and yield of B decrease while the hydrogen consumption and the yield of C go through a maximum with an increase in liquid velocity. These results are in qualitative agreement with the experimental results reported by Shah et al. (1978b).

f. Effect of column diameter: An increase in column diameter results in an increase in the backmixing of the three phases and a decrease in the gas holdup. The catalyst concentration gradient decreases throughout the reactor as the column diameter increases. The effect of this decrease in the local catalyst concentration on the reaction rates more than compensates for the effect of the increase in liquid-phase holdup on the reaction rates. As a result, all yields and consumptions as well as the fraction of liquid vaporized show a decrease with an increase in column diameter.

g. Effect of reactor length: Residence time as well as Peclet numbers for the gas and liquid phases increase with an increase in the reactor length. Parulekar (1979) found that these factors help to improve the yields of B and C. There is also an increase in the consumption of both A and hydrogen along with an increase in the total vaporization of liquid as the reactor length is increased. His results were in qualitative agreement with the regression plots for the yields of the various products as a function of reactor residence time for Kentucky coal obtained in pilot-plant (Fort Lewis, Gulf, Willsonville, etc.) operations (Jones (1979)).

h. Effect of weight fraction of solid in feed slurry: For catalyst loading of 15% to 50% wt, the effect of the increase in the local catalyst

concentration on all the reaction rates is more prominent than the effect of the decrease in the liquid holdup on the reaction rates. Therefore, all the yields and consumptions increase as W_s is increased. For a catalyst loading higher than 50 wt%, the effect of a decrease in the liquid holdup on reaction rates more than compensates for the effect of an increase in the local catalyst concentration. Hence, all yields and consumptions show a decrease with any further increase in W_s .

1. Effect of particle size: An increase in the particle size of the catalyst increases the effect of settling and thus increases the catalyst concentration gradient and the local catalyst concentration in the reactor as well as decreases the local liquid holdup. Parulekar (1979) showed that for particle sizes up to 275 μm , all the yields and consumptions increase as the particle size increases. Any further increase in particle size above 275 μm results in a reduction of all yields and consumptions owing to the increasing importance of the effect of the further decrease in the liquid holdup on the reaction rates.

Parulekar (1979) has studied typical axial profiles of the dimensionless concentration of A, B, C, and H_2 , the dimensionless velocities of gas, liquid, and slurry, the holdups of the gas, liquid, and solid phases, and catalyst concentration C_s . These studies show that significant axial variations in the superficial liquid and gas velocities and the holdups of various phases could be expected for certain operating conditions. However, the validity of these predictions has not been checked with actual experimental data.

Other Complex Reactor Models:

Lee et al. (1978) have proposed an isothermal reaction model which considers the gas-liquid mass transfer resistance. They assumed axially

constant superficial phase velocities and phase holdups for the gas and liquid phases. The model developed is applicable with any kinetic rate expression. They applied an axial dispersion model to the preheater reactor assembly of the SRC process and simulated the Wilsonville SRC-I pilot plant. A plug flow reactor model was assumed for the preheater whereas an axial dispersion model was assumed for the reactor. Coal dissolution, hydrodesulfurization and hydrogenation reactions were assumed as key reactions, and the kinetic rate expressions developed by Pitts (1976) were used in their analysis.

Lee et al. (1978) concluded that the Wilsonville SRC plant operates in the kinetically controlled regime. Singh et al. (1982) have theoretically analyzed the conditions which may lead to hydrogen starvation due to the low transfer rates of hydrogen to the liquid phase. Singh et al. (1982) have proved that the possibility of hydrogen starvation would be at a minimum for superficial gas velocities greater than 1 cm/sec. It seems that even if Singh et al. (1982) have used an erroneous correlation for the prediction of the mass transfer coefficient, their conclusion of insignificant mass transfer resistance would be valid for industrial reactors.

Other conclusions from the work of Lee et al. (1978) include: (a) coal dissolution is nearly complete in the preheater, and (b) the effect of gas and liquid backmixing on coal dissolution becomes more significant when the hydrogen mass transfer coefficient is low. For an industrial size reactor, the hydrogen mass transfer coefficient as well as the gas and liquid backmixing will always be high.

The application of models developed by Parulekar and Shah (1980) can be viewed as a typical application of the design procedure described by Ledakowicz et al. (1983). Any good model should be based on realistic assumptions backed by rough estimations. The model equations are usually

solved numerically as they contain strong non-linearities. The general scheme presented by Ledakowicz et al. (1983) must be iterated many times as the desired optimal conditions cannot be calculated explicitly, and it is usually subject to various economic considerations. In the context of SRC-II reactor scaleup problems, a major design question was whether to build one tall reactor or two reactors of equivalent height.

The use of models and simulations are extremely valuable in the design and scaleup considerations. Though highly sophisticated models can be formulated on the basis of first principles and can be solved using numerical methods, the design variables can only be meaningful to the extent of the accuracy of data.

The models developed by Parulekar and Shah (1980) and Lee et al. (1978) are highly sophisticated, and the use of these complex models is justified for experiments in small laboratory liquefaction units with high length to diameter ratios. For the case of industrial coal liquefaction reactors with larger diameters, simpler models might be more realistic to predict the general behavior of coal liquefaction reactors.

Ledakowicz et al. (1983) have studied the usefulness of simpler models in the scaleup considerations for industrial scale coal liquefaction reactors. On the basis of rough parameter estimations, Ledakowicz et al. (1983) found that for large scale industrial reactors the liquid phase as well as the gas phase can be approximated by CSTR behavior. In addition, the liquefaction process is entirely reaction controlled, and hence the multiphase reactor problem can be approximated by a pseudohomogenous reaction. Before making recommendations about use of any one model, efforts should be made to analyze the available data using various models.

Nomenclature:

A	Heavy distillate
A	Dimensionless concentration of heavy distillate as defined in equation 8
a	Liquid phase specific mass-transfer area at gas-liquid interface, m^{-1}
B	Light distillate
B	Dimensionless concentration of light distillate as defined in equation 8
C	Volatile products
C	Dimensionless concentration C as defined in equation 8
$C_{A\ell}$	Concentration of A, $kg/(m^3 \text{ liquid})$
$C_{B\ell}$	Concentration of B, $kg/(m^3 \text{ liquid})$
C_C	Concentration of C, $kg\text{-mol}/(m^3 \text{ liquid})$
C_{H_2}	Concentration of hydrogen, $kg\text{-mol}/(m^3 \text{ gas})$
C_s	Concentration of solids, $kg/(m^3 \text{ slurry})$
C_T	Total gas-phase molar concentration, $kg\text{-mol}/(m^3 \text{ gas})$
D_C	Diameter of column, m
D	Diffusivity, m^2/s
D_H	Axial thermal-dispersion coefficient, $kJ/(m\cdot s\cdot K)$
$D_{i\ell}$	Diffusivity of component i in liquid phase, m^2/s
d_{vs}	Sauter mean bubble diameter, m
e	Dimensionless mass dispersion coefficient
E_2	Axial mass-dispersion coefficient, m^2/s
$E_1\text{-}E_4$	Activation energies for reaction (1) through (4) respectively, $kJ/kg\text{-mol}$
f, f_1	Quantities defined by equation 8
g	Gravitational acceleration, m/s^2
H	Henry's-law constant, $[kg\text{-mol}/(m^3 \text{ liq.})]/[kg\text{-mol}/(m^3 \text{ gas})]$

H'	Dimensionless concentration of hydrogen
H_0	Static slurry height in column (m)
k_L	Liquid-side mass-transfer coefficient, m/s
k_{La}	Volumetric liquid-side mass-transfer coefficient, m/s
k_1, k_1^*	Kinetic constant and frequency factor for reaction (1) (m^3 slurry)/kg catalyst) (sec)
k_2, k_2^*	Kinetic constant and frequency factor for reaction (2) (m^3 gas) (m^3 slurry)/(sec)(kg catalyst) (kg A)
k_3, k_3^*	Kinetic constant, and frequency factor for reaction (3) (m^3 gas) (m^3 slurry)/(sec)(kg catalyst) (kg B)
k_4, k_4^*	Kinetic constant and frequency factor for reaction (4), (m^3 gas) (m^3 slurry)/(m^3 liq.)(kg catalyst) (sec)
L	Length of column (m)
M_{H_2}, M_C	Molecular weights of hydrogen and product gases (C) respectively, kg/kg-mol
n	Mass flux, kg/($m^2 \cdot s$)
P	Pressure, MPa
P'	Power consumption
Pe	Peclet number
r	Ratio of superficial liquid velocity to superficial gas velocity
R_g	Universal constant MPa. m^3 /(kg-mol.K) or (kJ/kg-mol.K)
Re_s	Particle Reynold's number defined in Table I-1
r_1	Rates of reaction (1), kg/(m^3 liq.) (sec)
r_2, r_3, r_4	Rate of reaction, (2), (3), (4), kg-mol/(m^3 liq.) (sec)
R_1-R_4	Quantities defined by Equation 8
T	Temperature, K
U	Net velocity of a phase, m/s
U	Dimensionless superficial velocity
$U_{b\infty}$	Bubble rise velocity, m/s

U_{og}, U_{ol}, U_s	Superficial velocity of gas, liquid, and solid phase respectively, m/s
U_t	Settling velocity with respect to liquid, m/s
U_t'	Terminal settling velocity of particles, m/s
W_s	Weight fraction of solid in feed slurry, kg/(kg slurry)
x	Distance from inlet of reactor, m
Y_A, Y_{H_2}	Consumption of heavy distillate and hydrogen respectively, kg/(kg feed slurry)
Y_B, Y_C	Yields of light distillate and volatile products respectively, kg/(kg feed slurry)
z	Dimensionless distance from the bottom of column

Greek Letters

$\epsilon_g, \epsilon_l, \epsilon_s$	Holdup of the gas, liquid and solid phase respectively
μ	Viscosity, kg/(m.s)
ν	Kinematic velocity, m ² /s
ρ	Density, kg/m ³
σ	Surface tension of liquid, kg/s ² or N/m

Subscripts

b	Bulk slurry
C	Volatile products
g	Gas phase
H_2	Hydrogen
l, L	Liquid phase
s	Solid phase
sl	Solid-liquid slurry
w	Wall
o	Feed condition
l	Reactor outlet condition

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Table I-1

Correlations for Estimation of Design Parameters for
Bubble Column Slurry Reactors

1. Bubble Size:
Calderbank (1967)

$$d_{vs} = C \frac{\sigma^{0.6}}{(P'/V_D)^{0.4} \rho_L^{0.2}} E_G^n \left[\frac{\mu}{\mu_L} \right]^{0.25}$$

2. Terminal Bubble Rise Velocity:

$$U_{b\infty} = \frac{\mu_L}{\rho_L d_{vs}} M^{-0.149} (J - 0.857)$$

where

$$M = \frac{g \mu_L^4 (\rho_L - \rho_G)}{\rho_L^2 \sigma^3}, \quad Re = \frac{d_{vs} U_{b\infty} \rho_L}{\mu_L}$$

$$E_o = \frac{g (\rho_L - \rho_G) d_{vs}^2}{\sigma}$$

$$J = 0.94 H^{0.747} \quad (2 < H < 59.3)$$

and $J = 3.42 H^{0.441} \quad (H > 59.3)$

where $H = \frac{4}{3} (E_o) M^{-0.149} \left(\frac{\mu_L}{\mu_w} \right)^{-0.14}$

for the range $M < 10^{-3}$, $E_o < 40$, $Re > 0.1$

3. Gas Holdup:
Akita and Yoshida (1973)

$$\frac{e_g}{(1-c_g)^4} = 0.2 \left[\frac{g D_c^2 \rho_L}{\sigma} \right]^{1/8} \left[\frac{g D_c^3}{v_L^2} \right]^{1/12} * \left[\frac{U_G}{\sqrt{g D_c}} \right]$$

4. Volumetric mass transfer coefficient:
Akita and Yoshida (1973)

$$\frac{k_L a D_c^2}{D_i} = 0.6 \left[\frac{v_l}{D_i} \right]^{0.5} \left[\frac{g D_c^2 \rho_l}{\mu_l^2} \right]^{0.62} \left[\frac{g D_c^3}{v_l^2} \right]^{0.31} \epsilon_g^{1.1}$$

5. Liquid phase dispersion coefficient.
Deckwer et al. (1974)

$$E_{zL} = 0.678 D_c^{1.4} U_g^{0.3}$$

Joshi (1980)

$$E_{zL} = 0.33 (U_c + U_l) D_c$$

where,

$$v_c = 1.31 \left[g D_c \left(U_g - \frac{\epsilon_g}{1-\epsilon_g} U_l - \epsilon_g U_{b\infty} \right) \right]^{1/3}$$

6. Gas phase dispersion coefficients
Mangartz and Pilhofer (1980)

$$E_{zG} = 50 D_c^{1.5} (U_g/\epsilon_g)^3$$

Field and Davidson (1980)

$$E_{zG} = 56.4 D_c^{1.33} (U_g/\epsilon_g)^{3.56}$$

7. Solid phase dispersion coefficient

$$\frac{U_g D_c}{E_{zs}} = Pe_s = \left[1 + 0.009 Re_s \left(\frac{U_g}{\sqrt{g D_c}} \right)^{-0.8} \right] \frac{13 (U_g / \sqrt{g D_c})}{1 + 8 (U_g / \sqrt{g D_c})^{0.85}}$$

$$\text{Where } Re_s = \frac{d_p U}{v_L}$$

Table I-2

Physical Properties and Kinetic Parameters (Parulekar, 1979)

(1) Frequency Factors:

k_1^*	$20 \frac{\text{m}^3 \text{ slurry}}{(\text{kg cat.}) (\text{sec})}$
k_2^*	$10 \frac{(\text{m}^3 \text{ gas}) (\text{m}^3 \text{ slurry})}{(\text{sec}) (\text{kg cat.}) (\text{kg A})}$
k_3^*	$50 \frac{(\text{m}^3 \text{ gas}) (\text{m}^3 \text{ slurry})}{(\text{sec}) (\text{kg cat.}) (\text{kg B})}$
k_4^*	$0.4 \frac{(\text{m}^3 \text{ gas}) (\text{m}^3 \text{ slurry})}{(\text{m}^3 \text{ liq.}) (\text{kg cat.}) (\text{sec})}$

(2) Activation Energies, kJ/kg-mol: .

E_1	8.29×10^4
E_2	1.16×10^5
E_3	5.86×10^4
E_4	1.24×10^5

(3) Solid Density ρ_s	$1.3 \times 10^3 \text{ kg/m}^3$
(4) Liquid Viscosity μ_g	$0.9 \times 10^{-2} \text{ Pa.s}$
(5) Interfacial Tension σ	$8 \times 10^{-3} \text{ N/m}$
(6) Universal Gas Constant (R_g)	$8.315 \times 10^3 \frac{\text{m}^3 \text{ Pa}}{\text{kg-mol K}}$
(7) Molecular weight, kg/kg-mol:	
Hydrogen	2
Volatile Products (C)	40
(8) Liquid Density ρ_l	$0.9 \times 10^3 \text{ kg/m}^3$
(9) Weight fraction of heavy oil in feed liquid, $A_o = 0.7$.	
Weight fraction of light oil in feed liquid, $B_o = 0.3$.	
Mole fraction of hydrogen in feed gas, $H_o = 1$.	

Table I-3

Operating Conditions and Reaction Dimensions (Parulekar, 1979)

(1)	Column (reactor) diameter	0.08m
(2)	Reactor length	5.4m
(3)	Reactor feed temperature	623 K
(4)	Total pressure	10.13 MPa
(5)	Weight fraction of catalyst in feed slurry, W_s	0.15
(6)	Particle size	150 μm
(7)	Superficial gas velocity $(U_{og})_o$	1.5×10^{-2} m/sec
(8)	Superficial liquid velocity $(U_{ol})_o$	1.5×10^{-3} m/sec