

**DESIGN OF SLURRY REACTOR
FOR INDIRECT LIQUEFACTION APPLICATIONS**

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by

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INTRODUCTION

Slurry bubble reactors are expected to be used for a number of applications which include production of methanol and hydrocarbon fuels via Fischer-Tropsch route using synthesis gas from coal gasification. These reactors can operate with low H_2/CO ratio without the possibility of plugging as a consequence of wax and/or carbon buildup. In addition these reactors also offer the advantages of high heat transfer rate, isothermal conditions of operation, reduced rate of catalyst deactivation due to washing effect of the liquid and on-line catalyst addition and withdrawal.

The objective of this project is to design and model a conceptual slurry reactor for two indirect liquefaction applications; 1) production of methanol and 2) production of hydrocarbon fuels via Fischer-Tropsch route. The reactor models for the two processes have been formulated using computer simulation. Process data, kinetic and thermodynamic data, heat and mass transfer data and hydrodynamic data have been used in the mathematical models to describe the slurry reactor for each of the two processes. For task 1, a mathematical model for methanol synthesis was developed and tested. This paper presents the mathematical model and technical approach used to design and model the slurry reactor for Fischer-Tropsch synthesis.

Fischer-Tropsch synthesis is essentially hydrogenation of carbon monoxide which yield saturated and unsaturated compounds of the homologous series. It permits the synthesis of hydrocarbons ranging from methane to high melting paraffins depending on the catalyst, temperature and type of process employed. A small quantity of byproducts such as alcohols, aldehydes, ketones etc. are also formed.

Fischer-Tropsch Slurry Reactor:

The problem of reactor design and modeling problem for Fischer-Tropsch synthesis has been divided into two parts.

- 1) Estimation of syn gas conversion in a given reactor volume.
- 2) Detailed calculation of compositions of vapor and liquid product streams based on phase equilibria.

Reactor Model

The development of a reactor model starts with writing the differential mass and energy balance equations. The resulting differential equations can then be solved using suitable boundary conditions. Given the large number of products formed during FT synthesis, it is not practical to make individual component balance. For design purposes, however, some products can be grouped together without significant loss of accuracy. The reactor design configuration and its operating conditions also allow for some simplifying assumptions to be made.

The following assumptions were made for the development of FT synthesis reactor model.

Assumptions for FT reactor model:

- 1) The synthesis products obey Schulz-Flory distribution. This assumption has been found to be

valid over the entire range of products of an iron catalyst (Satterfield and Huff, 1982).

$$p_n = \alpha p_{n-1} = \alpha^{n-1} p_1 \quad (1)$$

Here, p_n refers to the product distribution as formed on the catalyst. The probability of chain growth α is taken to be constant over the entire carbon distribution.

2) The hydrocarbon products consist of n-alkanes and n-alkenes only. These are in fact the main products of catalysts and little design error is introduced by lumping the methyl branched isomers with n-alkenes and n-alkanes.

3) The gas phase component balance is made only for i) CO, ii) H₂, iii) CO₂, iv) H₂O and v) C₁ to C₃ hydrocarbons. The contribution of hydrocarbons heavier than C₃ is estimated to be less than 4% of the gas flow rate in the reactor.

4) The effectiveness factors for the catalyst particles are taken as unity. Due to the relatively small particle sizes (< 50 μ m) used in the reactors, diffusional limitations should be negligible.

5) Again, as a consequence of the small particle size, mass and heat transfer resistances between the catalyst and liquid are assumed to be negligible.

6) The gas and slurry phases are modeled using the axial dispersion model, the most appropriate

model for bubble column reactors (Deckwer et al., 1983).

7) The catalyst is not uniformly distributed in the reactor and the sedimentation-dispersion model (Cova, 1966; Kato et al., 1972, Smith et.al, 1985) is used for modeling the catalyst concentration.

8) The hydrodynamic parameters, namely gas holdup, interfacial area, heat and mass transfer coefficients and dispersion coefficients are assumed to be spatially independent.

9) The temperature dependence of the gas phase concentration $C_G = P/RT$ can be neglected since

$$d(1/T)dz = -1/T^2(dT/dz) \approx 0$$

10) Steady-state conditions prevail

Model Equations:

Based on the above assumptions, mass and energy balance were developed for the reactor model. Component balance were made for the following components. Definitions of the variables, coefficients and dimensionless groups are given in the Notation.

- 1) CO
- 2) H₂
- 3) CO₂
- 4) H₂O
- 5) C₁ to C₃

Gas Phase Mass Balance:

The gas phase mass balance with axial dispersion for component i can be written as:

$$\frac{1}{Pe_G} \frac{d^2 y_i}{dz^2} - \frac{d(vy_i)}{dz} - (St_{G,i})(y_i - x_i) = 0 \quad (2)$$

subject to the following boundary conditions:

$$vy_i - \frac{1}{Pe_G} \frac{dy_i}{dz} = y_{i,0} \quad \text{at } z=0 \quad (3)$$

$$\frac{dy_i}{dz} = 0 \quad \text{at } z=1 \quad (4)$$

The molar flow rate of gas will vary along the column due to reaction. The effect of change in molar flow rate on the superficial gas velocity can be determined by writing an overall mass balance. Thus

$$-\frac{dv}{dz} - \sum St_{G,i}(y_i - x_i) = 0 \quad (5)$$

The above equation is subject to the boundary condition

$$v = 1 \quad \text{at } z = 0 \quad (6)$$

The volumetric flow rate of the gas will also vary along the column due to change in hydrostatic head. The local pressure along the column is given as

$$P(z) = P_T [1 + \beta(1-z)] \quad (7)$$

where

$$\beta = \rho_{sl}g(1-\epsilon_g)L/P_T \quad (8)$$

Liquid Phase Mass Balance:

The liquid phase mass balance for component i can be written as:

$$\frac{1}{Pe_L} \frac{d^2x_i}{dz^2} - \frac{dx_i}{dz} + St_{L,i}(y_i - x_i) + \sum \theta_{i,k}(1-\epsilon_g)C_{cat}r_k(H_iL/PU_L) = 0 \quad (9)$$

subject to the following boundary conditions:

$$- \frac{1}{Pe_L} \frac{dx_i}{dz} + x_i = x_{i,o} \quad \text{at } z=0 \quad (10)$$

$$dx_i/dz = 0 \quad \text{at } z = 1 \quad (11)$$

Solid Phase Mass Balance:

The axial distribution of catalyst is governed by gravitational settling and axial dispersion due to agitation of the catalyst slurry by the gas flow. From assumption (8) the volume fraction of the liquid is constant along the column length and hence the mass balance on catalyst can be written as:

$$\frac{1}{Pe_s} \frac{d^2w}{dz^2} + \frac{dw}{dz} = 0 \quad (12)$$

with boundary conditions:

$$w - 1/Pe_z(dw/dz) = 0 \quad (13)$$

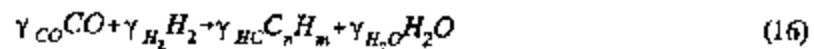
$$\int_0^1 w dz = 1 \quad (14)$$

An analytical solution to the above equation is possible and is given below:

$$w = \frac{Pe_z \exp(-Pe_z z)}{[1 - \exp(-Pe_z)]} \quad (15)$$

Stoichiometry of Fischer-Tropsch reaction:

The general stoichiometry of Fischer-Tropsch reaction can be given as:



Water formed as primary product is converted to carbon dioxide by the water gas shift reaction in presence of iron catalyst.



As shown by Stern et al. (1985), the stoichiometric coefficients (γ_i) and average number of carbon (n) and hydrogen (m) atoms in the hydrocarbon products can be expressed in terms of the probability of chain growth (α) and the fraction of paraffinic hydrocarbons (λ) variables.

$$Y_{CO} = \frac{1}{D} \quad (18)$$

$$Y_{H_2} = \frac{2 + (1 - \alpha)^2 + \lambda \alpha (1 - \alpha)}{D} \quad (19)$$

$$Y_{HC} = \frac{(1 - \alpha)}{D} \quad (20)$$

$$Y_{H_2O} = \frac{1}{D} \quad (21)$$

$$D = 3 + (1 - \alpha)^2 + \lambda \alpha (1 - \alpha) \quad (22)$$

$$n = (1 - \alpha)^{-1} \quad (23)$$

$$m = 2[(1 - \alpha)^{-1} + (1 - \alpha) + \lambda \alpha] \quad (24)$$

The value of chain growth probability factor (α) and fraction of paraffins in the product (λ) are a function of the catalyst and operating conditions. For the production of high molecular weight products, value of α has been found to be in the range of 0.85-0.95.

Kinetics of Fischer-Tropsch Synthesis:

The reaction kinetics of Fischer-Tropsch synthesis over iron catalyst have been investigated by a number of researchers. The various kinetic expressions have been reviewed by Huff and Satterfield

(1984) and Zimmerman and Bukur (1990). The rate expression proposed by Anderson (1956) was found to be particularly useful.

$$r_{FT} = \frac{k_0 P_{CO} P_{H_2}}{P_{CO} + a_1 P_{H_2O}} \quad (25)$$

This equation includes water inhibition effect. At synthesis gas conversion below about 60 % Equation (25) reduces to

$$r_{FT} = k_1 P_{H_2} \quad (26)$$

Huff and Satterfield (1984) derived an alternate form of expression based on the observed hydrogen dependance for a fused-magnetite catalyst.

$$r_{FT} = \frac{k_2 P_{CO} P_{H_2}^2}{P_{CO} P_{H_2} + b_1 P_{H_2O}} \quad (27)$$

Equations (25) and (27) have the same form if the constant 'a₁' in Equation (25) depends on the H₂ partial pressure i.e. a₁ = b₁/H₂.

For precipitated iron catalyst (100Fe/0.3Cu/0.2K), Zimmerman and Bukur (1990) observed that Equation (25) gave a better fit to their data compared to Equation (27). Similar observations were made by Nettelhoff et al. (1985) using a precipitated, unpromoted iron catalyst. Based on these observations, kinetic expression given by Equation (25) was selected for precipitated iron catalyst in the FT reactor model. The rate constants and adsorption constants were obtained from experimental data of Zimmerman and Bukur (1990).

Water gas shift reaction:

The water gas shift reaction occurs readily over potassium promoted iron catalyst. The shift reaction enables CO rich feeds to be utilized efficiently without the need for external shift. Both Equations (16) and (17) for the FT and WGS reaction must be considered to accurately predict both H₂+CO conversions and H₂/CO usage ratio which requires knowledge of WGS kinetics. The following rate expression for water gas shift reaction has been selected based on literature information (Kuo, 1983; Zimmerman and Bukur, 1990).

$$r_{WGS} = k_{wgs} \frac{(P_{CO} P_{H_2O} - P_{CO_2} P_{H_2} / K_p)}{P_{CO} + a_1 P_{H_2O}} \quad (28)$$

It may be pointed out here that literature kinetic expressions for FT synthesis are based on partial pressures in the gas phase while in a slurry reactor the rate depends on the liquid phase concentrations. Zimmerman and Bukur (1990) pointed out that for FT synthesis in a well mixed slurry reactor (i.e. stirred autoclaves), the partial pressures and liquid concentrations can be related by assuming ideal gas phase and Henry's law behavior in the liquid phase.

$$P_i = H_i C_{L,i} \quad (29)$$

Kinetic expressions based on liquid phase concentrations were obtained using Equation (29).

It may be noted, however, that the underlying assumption of gas and liquid phase equilibrium may not be justified for fast reaction systems. Therefore kinetic expressions based on actual liquid phase concentrations need to be developed.

Estimation of Model Parameters:

Gas Holdup

Recently Bukur et al. (1990) reported hydrodynamics of three-phase slurry Fischer-Tropsch bubble column reactors. Experiments were conducted in both batch bubble column mode and continuous bubble column mode to investigate the effects of solids concentration (0 to 30 wt. %), solid particles type and size (iron oxide and silica particles; 0-5 μm and 22-44 μm). Two types of liquids, namely hydrotreated reactor wax (FT-300) and SASOL reactor wax were used. The operating conditions were generally selected to closely simulate slurry bubble column reactors for Fischer-Tropsch synthesis.

Bukur et al. (1990) tested various literature correlations using their experimental data. It was found that correlation of Zheng et al. (1988), Badjugar et al. (1986) and Hughmark (1967) could provide reasonable estimates of gas holdups in slug flow and churn turbulent regimes. It may be pointed out that we selected the correlation of Hughmark (1967) for methanol synthesis reactor simulation based on a test of various literature correlations using the experimental data of Air Products PDU operation (Prakash and Bendale, 1990). Based on the observation of Bukur et al. (1990), the correlation of Hughmark (1967) could also be used for a Fischer-Tropsch reactor model. Bukur et al. (1990) also developed a general correlation for gas holdup by combining the data of two-phase studies (Bukur et al., 1987a,b) and three-phase studies. This correlation is recommended by the authors for estimation of gas holdups in Fischer-tropsch slurry bubble column reactors operating in slug flow and churn turbulent regime

$$c_G = 0.24 Fr_G^{0.28} Bo^{0.14} \quad (30)$$

This correlation has also been included in the slurry reactor model.

Volumetric Mass Transfer Coeff.

The correlation of Nguyen-tien et al. (1985) as modified by Nigam and Schumpe (1987) was selected for the estimation of volumetric mass transfer coefficient. The proposed correlation is applicable to both continuous and batch operated slurry bubble columns.

$$k_{La} = 0.39(1 - \epsilon_{s,max})^{0.58} U_g^{0.67} \quad (31)$$

Nigam and Schumpe (1987), however, observed that for slurry bubble columns with batch operation of the liquid phase, the mean solid fraction ($\epsilon_{s,max}$) should be replaced by solid fraction at the reactor bottom ($\epsilon_{s,b}$).

Although the correlation of Nguyen-tien et al. (1985) accounts for the effects of solids concentration and gas velocity, it is based on data obtained for oxygen mass transfer in aqueous solutions. For other systems, it needs to be corrected for the effects of diffusivities, physical properties of liquid and coalescing behavior of the system.

Correction for Diffusivity:

Correlations for volumetric mass transfer in bubble columns (Akita and Yoshida, 1973; Ozturk et al., 1987) show that:

$$k_{L_v} \propto D_i^{0.5} \quad (32)$$

This assumes that gas-liquid mass transfer in bubble columns is governed by Higbie's penetration theory.

Correction for Physical Properties of Liquid and Slurry:

Volumetric mass transfer coefficient in bubble columns has been found to be affected by viscosity, surface tension and density of the liquid medium (Akita and Yoshida, 1973; Hikita et al., 1981; Ozturk et al., 1987). Akita and Yoshida (1973) and Hikita et al. (1981) studied gas-liquid mass transfer in aqueous solutions, while Ozturk et al. (1987) used organic liquids for their study. The effects of changes in surface tension can be enhanced in aqueous solutions of oxygenated compounds (i.e. alcohols) due to a simultaneous change in the coalescing behavior of liquid. The correlation of Ozturk et al. (1987) was selected to apply corrections for physical properties of liquid. It gives:

$$(k_{L_v})_1 \propto \sigma_L^{-0.33} \mu_L^{-0.08} \rho_L^{0.37} \quad (33)$$

It may be noted that the correlation of Ozturk et al. (1987) is based on data obtained in solid-free bubble column. The surface tension effects are not expected to change in presence of solids. The

effect of slurry viscosity is accounted for by the term in the brackets in the correlation of Nguyen-tien et al. (1985). For density correction in slurry bubble columns, the liquid density can be replaced by slurry density, since fine particles in slurry reactor form a homogeneous slurry phase. The correlation of Nguyen-tien et al. (1985) can, therefore, be corrected as below to estimate volumetric mass transfer coefficients in other systems:

$$(k_L a)_s = (k_L a)_o (D_s/D_{o,w})^{0.5} (\sigma_s/\sigma_w)^{-0.33} (\mu_s/\mu_w)^{-0.08} (\rho_{sl}/\rho_w)^{0.37} \quad (34)$$

The above correlation is applicable to coalescing media, for noncoalescing media (and other high gas holdup conditions) it would give conservative estimates of volumetric mass transfer coefficient.

Liquid Phase Dispersion Coefficient:

While liquid phase dispersion in solid-free bubble columns has been investigated extensively, little work has been done to measure liquid backmixing in slurry bubble columns. However, in the suspension of fine catalyst particles used in Fischer-Tropsch synthesis reactors, there is expected to be little effect of the presence of solids on liquid mixing. The various correlations available in the literature for liquid phase dispersion in bubble column were compared by Wendt et al. (1984). The correlation proposed by Deckwer et al. (1974) provided a good estimate of the liquid phase dispersion coefficient. This correlation was, therefore, selected to estimate the liquid phase dispersion coefficient in the slurry reactor.

Deckwer et al. (1974)

$$D_L = 0.68 D_C^{1.4} U_G^{0.3} \quad (35)$$

Gas Phase Dispersion Coefficient:

Earlier we had selected the correlation of Towell and Ackerman (1972) to estimate the gas phase axial dispersion coefficient in bubble columns (Prakash and Bendale, 1990). This correlation was found to provide good estimates of overall gas phase dispersion coefficients for various literature studies (Kawagoe et al., 1989). A recent report from Air Products and Chemicals (Topical Report: Tracer Studies in the LaPorte LPMEOH PDU, 1990), however, showed that literature correlations (Towell and Ackerman, 1972; Field and Davidson, 1980) over predicted gas phase mixing by a factor of about three in their large diameter slurry bubble column reactor. A new correlation was then recommended based on the best fit of their data.

Towell and Ackerman (1972)

$$D_G = 20.0 D_C^2 U_G \quad (36)$$

Air Products and Chemicals (1990)

$$D_G = 21.7 D_C^{1.5} U_G^{1.8} \quad (37)$$

Axial Solids Distribution:

Although several groups of researchers have investigated distribution of solids in slurry bubble columns, the work of Bukur et al. (1989), using Fischer-Tropsch waxes, is more relevant for our purposes. Bukur et al. (1989) observed that the axial distribution for the 0-5 μm and 20-44 μm iron oxide and silica particles were uniform for all runs in the continuous mode of operation. For batch mode of operation, the 0-5 μm particles showed only a slight gradient with high concentration towards the bottom of the column. However, with 20-44 μm particles significant gradients in axial solids distribution profiles were observed in the batch mode of operation. Bukur et al. (1989) also noted that the correlation of Smith and Ruether (1985) provided a good prediction of solids dispersion coefficient in slurry bubble columns.

Smith and Ruether (1985)

$$Pe_p = 9.6(Fr_G^6 / Re_G)^{0.116} + 0.019 Re_p^{1.1} \quad (38)$$

$$Re_p = Ar/18 \quad \text{if } Re_p < 0.05 \quad (39)$$

$$Re_p = (Ar/13.9)^{0.7} \quad \text{if } Re_p > 0.5 \quad (40)$$

Smith and Ruether (1985) have suggested the following equation for hindered settling velocity in particle swarm.

$$U_s = 1.1 U_G^{0.026} U_r^{0.8} \epsilon_L^{3.5} \quad (41)$$

Physical, Thermodynamic and Transport Properties:

Various literature correlations and models were reviewed to select suitable methods for estimation of physical, thermodynamic and transport properties. Whenever possible, available experimental data was used to develop suitable correlations.

Diffusivities:

Akgerman measured the diffusion coefficients for synthesis gas in high molecular weight liquids ($n\text{-C}_{20}$ and $n\text{-C}_{28}$) and FT waxes for a DOE contract DE-AC22-84PC 70032 at temperatures ranging from 373 to 534°K. The following correlation was proposed to estimate diffusion coefficients.

$$\frac{10^9 D_i}{T^{0.5}} = \frac{94.5(V_s - c_i V_{so})}{M_i^{0.239} M_s^{0.781} (\xi_i \xi_s)^{1.134}} \quad (42)$$

Here

$$c_i V_{so} = \frac{c_i N \xi_s^3}{\sqrt{2}} \quad (43)$$

$$c_i = 1.206 + 0.0632(\xi_i / \xi_s) \quad (44)$$

Henry Law Constants:

Chao and Lin (1987) investigated the solubilities of synthesis gas in high molecular weight solvents ($n\text{-C}_{20}$, $n\text{-C}_{28}$ and $n\text{-C}_{36}$) and Fischer-Tropsch waxes for a DOE contract No. DE-AC22-84PC 70024. Solubilities of hydrogen, carbon monoxide, methane and ethylene were measured at temperatures in the range of 100-300°C and pressure 10-50 atm. Lee (1986) studied phase equilibria for syngas in Witco-40 and Freezene-100 oils. Graff et al. (1988) measured solubility of syngas in the temperature range of 210 to 260°C in another high molecular weight solvent (squalene). These authors observed that the Henry's coefficients could be well approximated by an equation of the form:

$$H_i = A_i \exp(B_i/T) \quad (45)$$

The solubility data of Chao and Lin (1987) in Fischer-Tropsch waxes was used to obtain the coefficients in above equation for different components.

Vapor-Liquid Equilibrium Calculations in Fischer-Tropsch Synthesis:

The major constituents of the products from Fischer-Tropsch synthesis are the hydrocarbons ranging from methane to high melting paraffins with relatively small quantities of olefinic compounds. For iron catalyst, carbon dioxide is one of the major products formed with relatively small amounts of product water. In the case of cobalt catalyst, however, due to its poor activity for water gas shift reaction, water will be the major byproduct with relatively small amount of carbon dioxide. Small quantities of other byproducts such as alcohols, aldehydes, ketones etc. are also formed. Straight-chain paraffins along with some 2-methylated branched paraffins predominate among the saturated hydrocarbons; major olefins are terminal olefins.

The products and the unreacted synthesis gas leaving the top of the slurry reactor will be assumed to be in equilibrium with the liquid phase in the reactor. A computer program has been developed to study the phase equilibria of the vapor and the liquid streams leaving the slurry reactor. Such information will be helpful in determining the relative flow rates and the composition of vapor and liquid streams which can be further used in the process design of the downstream units in the Fischer-Tropsch Plant. The conversion and the production yields from the slurry reactor model will be used to determine the overall composition for the phase equilibrium calculations. The problem presented here is identical with that of determining vapor and liquid compositions in a multi-component flash separation. Following assumptions will be made regarding the products formed during the reaction. The hydrocarbon products consists of only n-alkanes and n-alkenes. These are, in fact, the main products for many of the FT catalyst and little design error will be introduced by lumping the methyl-branched isomers with n-alkenes and n-alkanes. In the lower

oxygenates, ketones and primary alcohols are formed in relatively small amounts as compared to water and hence these compounds will be lumped with water. The light hydrocarbons (C_5 to C_{11}) will be lumped together; heavy hydrocarbons (C_{12} to C_{15}) will be lumped together and hydrocarbons C_{16} and heavier will be considered as the slurry reactor wax with an average composition of C_{40} paraffin. The olefin to paraffin split of the hydrocarbon products can be obtained from the literature. For example, unreacted synthesis gas and reaction products from the Mobil's pilot plant for Fischer-Tropsch synthesis may be characterized into following fourteen components:

<u>Component No.</u>	<u>Component(s)</u>
1	Carbon Dioxide
2	Water + Ketones & Primary Alcohols (Acetone & 1-Propanol)
3	Hydrogen
4	Carbon Monoxide
5	Methane
6	Ethylene
7	Ethane
8	Propylene
9	n-Propane
10	Butylene
11	n-Butane
12	C_5 to C_{11} Light Hydrocarbons Lumped as n-Octane
13	C_{12} to C_{15} Heavy Hydrocarbons as n-Tridecane
14	C_{16} and Heavier as Slurry Reactor Wax (C_{40} Paraffin)

Phase Equilibrium Calculations for Multicomponent Vapor-Liquid Phase Equilibria:

A technique has been presented by Bendale, 1991, to predict multicomponent vapor-liquid equilibria from the optimized binary interaction parameters obtained from the Peng Robinson equation of state, which is given as:

$$P = \frac{RT}{(V - b_{mix})} - \frac{a_{mix}}{V(V + b_{mix}) + b_{mix}(V - b_{mix})} \quad (46)$$

For a multicomponent mixture, the parameters a_{mix} and b_{mix} are given by the following expressions:

$$a_{mix} = \sum_i \sum_j x_i x_j \sqrt{a_i a_j} (1 - \delta_{ij}) \quad (47)$$

$$b_{mix} = \sum_i x_i b_i \quad (48)$$

In the above equation, δ_{ij} is defined as an interaction parameter that describes the deviation of parameter a_{mix} from the geometric mean of the pure component parameters a_i and a_j and is assumed to be constant. The pure component parameters a_i and b_i are given as:

$$a_i = a(T_r) \alpha(T_r, \omega) \quad (49)$$

$$a(T_r) = 0.45724 (R^2 T_c^2 / P_c) \quad (50)$$

$$\alpha(T_r, \omega) = [1 + \kappa (1 - \sqrt{T_r})]^2 \quad (51)$$

$$\kappa = C_a (0.37464 + 1.54226 \omega - 0.26992 \omega^2) \quad (52)$$

If ω is greater than 0.5, then

The optimized values of correction factors C_a and C_b for pure components are evaluated which

$$\kappa = C_{\kappa} (0.379642 + 1.48503 \omega - 0.164423 \omega^2 + 0.016666 \omega^3) \quad (53)$$

$$b_i = C_b [0.0778 (RT_c/P_c)] \quad (54)$$

minimize the sum of absolute relative errors of calculated and experimental saturated vapor pressure and saturated liquid density. The values of these correction factors approach unity for small molecules and non-polar gases which the Peng Robinson equation of state is known to model accurately. If these values are not available, then the default values are assigned as unity.

The expression for the fugacity coefficient obtained from the evaluation of the following equation,

$$RT \ln \Phi_i = \int_V^{\infty} \left[\left(\frac{\partial P}{\partial n_i} \right) - \frac{RT}{V} \right] dV - RT \ln Z \quad (55)$$

is given as:

$$\ln \Phi_i = \frac{b_i}{b_{mix}} (Z - 1) \cdot \ln (Z - B) + \left[\frac{\sum_k x_k (a_{ik} + a_{ki})}{a_{mix}} - \frac{b_i}{b_{mix}} \right] \times \frac{a_{mix}}{2.414 b RT} \ln \left[\frac{Z - 0.414 B}{Z + 2.414 B} \right] \quad (56)$$

$$B = \frac{b_{mix} P}{RT} \quad (57)$$

Phase equilibrium calculations are performed at constant temperature and pressure and known overall composition to determine the flow rates and compositions of vapor and liquid streams. The governing equations include overall and component material balances, mole fraction constraints, and thermodynamic equilibrium criterion of equal fugacities of each component in each phase. For an N component system at constant temperature and pressure, there will be 2N independent expressions for these components equilibrated in two phases, with 2N unknowns, i.e., V, x_i's and y_i's, respectively.

On the basis of one mole of mixture F (unreacted reactants and products formed), an overall material balance and a component balance for each component can be represented as follows:

$$L + V = F = 1 \quad (58)$$

$$Lx_i + Vy_i = Fz_i = z_i, \quad i = 1 \text{ to } N \quad (59)$$

with the following constraints:

$$\sum_i z_i = \sum_i x_i = \sum_i y_i = 1, \quad i = 1 \text{ to } N \quad (60)$$

and thermodynamic criteria,

$$f_i^V = f_i^L \quad \text{or} \quad \phi_i^V y_i P = \phi_i^L x_i P \quad \text{or} \quad \phi_i^V y_i = \phi_i^L x_i, \quad i = 1 \text{ to } N \quad (61)$$

To perform Flash calculations, temperature T and pressure P are considered as the known variables, which are used to calculate the unknown mole fractions x_i 's and y_i 's of the liquid and vapor phases, respectively. In addition to system temperature and pressure, the input data required consists of critical temperature, critical pressure and acentric factor of each component as well as the optimized binary interaction parameters, δ_{ij} as applied to the Peng-Robinson equation of state. The liquid and vapor phase fugacity coefficients for each component can be readily calculated from the expression 56. The set of equations 58, 59, 60 and 61 are solved simultaneously to determine the flow rates and mole fractions of liquid and vapor streams.

Development of Computer Codes:

The model equations for the Fischer-Tropsch synthesis reactor constitute a set of coupled second-order non-linear differential equations. These equations are not amenable to an analytical solution and therefore, a numerical method was selected for solution. Orthogonal collocation techniques are particularly suitable for the solution of boundary value problems and the software package, COLSYS, developed by Ascher et al. (1981) was selected for the numerical solution of the model equations. This method is based on spline collocation at Gaussian points using a B-spline basis. Approximate solutions are computed on a sequence of automatically selected meshes until a user-specified set of tolerances is satisfied.

Computer codes for the reactor model have been developed with a modular approach to computer programming, to ensure easy modifications by the user. Standard FORTRAN 77 has been used for writing the codes since this will ensure transfer to other compatible systems. Computer codes for the slurry reactor model have been developed for the following cases:

With external recirculation of slurry

- o Gas plug flow; Liquid axial dispersion
- o Gas axial dispersion; Liquid axial dispersion

No external recirculation of slurry

- o Gas plug flow; Liquid axial dispersion
- o Gas axial dispersion; Liquid axial dispersion

Results of Computation:

For the results of this section, the slurry reactor for Fischer-Tropsch synthesis was modeled assuming there was no external recirculation of slurry and both gas and liquid phases were axially dispersed. Table 1 gives the range of operating variables studied. The model was used to investigate the following:

- o Concentration profile along the reactor length
- o Effect of operating variables
- o Parameter sensitivity analysis
- o Simulation of demonstration unit

Table 1. Range of input data used to simulate slurry Fischer-Tropsch reactor

Diameter	4.5 m
Length	12.0 m
Temperature	230-270 °C
Pressure	15-20 atm
Gas Velocity	0.10-0.16 m/s
Slurry Conc.	30-35 wt. %
Syngas in Feed	90.0%
H ₂ /CO ratio	0.5-1.5
Particle size	0.00003-0.00005 m

Figure 1 shows how the concentration profile varied along the reactor length in both gas and liquid phases. It can be seen that in both phases the CO concentration declines more rapidly compared to hydrogen concentration as a result of the stoichiometry of the synthesis reaction. It may also be noted that the H₂/CO ratio in the liquid phase is higher than that in the gas phase.

Figures 2 and 3 show the effects of reactor temperature and gas velocity respectively, on syngas conversion. The conversion of syngas increased with increasing reactor temperature while it decreased with increasing gas velocity. Figure 3 also shows the results obtained with the gas plug flow case. It can be seen that predicted conversions were always higher for the gas plug flow mode than the gas dispersion mode and the difference between the two decreased with decreasing gas velocity. Figure 4 shows the effect of H₂/CO ratio in the feed gas on the conversion of syngas and CO. It can be seen that conversion of syngas passes through a maximum with increasing H₂/CO ratio.

Parameter sensitivity analysis for the reactor model was investigated for gas dispersion coefficient and volumetric mass transfer coefficient. Figure 5 shows that, as expected, the conversion decreases with increasing gas phase dispersion coefficient. The gas dispersion coefficient estimated by the selected correlation in the model is also shown on Figure 5. A 50% error in the estimated value of gas dispersion coefficient would result in less than 2% error in the predictions for syngas conversion. Figure 6 shows the significance of proper estimates for volumetric mass transfer coefficient in the model. It can be seen that syngas conversion would be significantly reduced for low gas-liquid mass transfer rates ($k_{1a} < 0.4 \text{ s}^{-1}$). The effect, however, becomes less significant for higher values of mass transfer coefficients.

Simulation of Demonstration Unit:

The Fischer-Tropsch reactor model was also used to predict the performance of demonstration unit at Rheinpreussen. Table 2 presents the reactor dimensions and operating conditions used. The predicted syngas conversion was 12 % higher than reported conversion. This could be attributed to higher catalyst activity at the Rheinpreussen plant.

Table 2. Operating conditions and reactor dimensions for Rheinpreussen
Demonstration Unit

Diameter	1.29 m
Length	7.7 m
Temperature	268°C
Pressure	11.84 atm
Gas velocity	0.095 m/s
Slurry Conc.	18 wt %

Syngas Conversion

Reported	Predicted
89%	77%

Simulation with Bechtel Design Data:

Bechtel presented a design for a commercial size slurry reactor for Fischer-Tropsch synthesis (Fox and Degen, 1990). Our slurry reactor model was used to predict the performance using Bechtel design data. The predicted syngas conversion was within 1% of reported value (Table 3).

Table 3. Bechtel Design Data

Diameter	4.8 m
Length	12.0 m
Net xsect of reactor	15.16 m ²
Reactor volume	211.0 m ³
Temperature	257°C
Pressure	28.3 atm
Slurry Conc.	35 wt%
Gas velocity	0.14 m/s

Syngas Conversion

Bechtel Design	Model Prediction
80%	79%

Conclusions and Recommendations:

- o A working computer model for Fischer-Tropsch synthesis in slurry reactor is available.
- o The model provide reasonable predictions for the demonstration unit.

There is a lack of information in the literature for the effect of following on reactor hydrodynamics:

- o decreasing gas velocity
- o presence of internals (i.e heat transfer tubes)
- o effect of operating pressure on gas holdup

Kinetic expressions based on actual liquid phase concentrations need to be developed.

Future Plan of Action:

The reactor model will be updated to include:

- o Kinetic model for cobalt catalyst
- o Calculations for heat transfer area

In addition we plan to present a design for gas distributor in the slurry reactor and provide preliminary cost estimates for commercial size reactors.

NOMENCLATURE

a_i	water adsorption coefficient, Equations (25),(28)
a_{mix}	Peng-Robinson attractive parameter for mixture [atm cm ⁶ / gmole ²]
a_i	Peng-Robinson attractive parameter for component i [atm cm ⁶ / gmole ²]
b_i	water adsorption coefficient, Equation (27), atm
b_{mix}	Peng-Robinson repulsive parameter for mixture [cm ³ / gmole]
b_i	Peng-Robinson repulsive parameter for component i [cm ³ / gmole]
c_i	coefficient in Equation 42, a function of molecular size of solute and solvent
C_1, C_2	Correction factors in the Peng-Robinson EOS
C_{cat}	catalyst concentration, kg/m ³ slurry
$C_{c,avg}$	average catalyst concentration, kg/m ³ slurry
$C_{G,i}$	gas-phase concentration of component i, kmol/m ³
$C_{L,i}$	liquid-phase concentration of component i, kmol/m ³
C_G	total gas phase concentration, kmol/m ³
C_p	heat capacity, kJ/kg.K
D_c	column diameter, m
D_G	gas-phase dispersion coefficient, m ² /s
D_i	diffusivity of component i in liquid phase, m ² /s
D_L	liquid-phase dispersion coefficient, m ² /s
$D_{o,w}$	diffusivity of oxygen in water, m ² /s
D_s	solid-phase dispersion coefficient, m ² /s
F	Moles in feed in flash calculations

f_i^v	Fugacity of component i in vapor phase [atm]
f_i^L	Fugacity of component i in liquid phase [atm]
H_i	Henry's constant for component i , atm.m ³ /kmol
k_0	rate constant for FT synthesis rate by Equation (25), kmol/kg-cat.s.atm
k_1	rate constant for first order FT synthesis rate by Equation (26), kmol/kg-cat.s.atm
k_2	rate constant for FT synthesis rate by Equation (27), kmol/kg-cat.s.atm
$k_{L,i}$	liquid-side mass transfer coefficient for component i , s ⁻¹
k_{w_0}	rate constant for water gas shift reaction by Equation (28), kmol/kg-cat.s.atm
L	reactor length, m in reactor model or moles of liquid phase in flash calculations
m	average number of hydrogen atoms in the hydrocarbon products
M_i	molecular weight of diffusing component i (kg/kgmol)
M_s	molecular weight of solvent (kg/kgmol)
n	average number of carbon atoms in the hydrocarbon products
N	Avogadro number
p_n	mole fraction of hydrocarbon products with carbon number n
P	Reaction Pressure [atm]
P_c	Critical pressure [atm]
P_i	partial pressure of component i , atm ($i = \text{CO}, \text{H}_2, \text{CO}_2, \text{H}_2\text{O}$)
$P_{r,i}$	Reduced pressure of component i
P_T	pressure at reactor top, atm
R	universal gas constant, 0.082 m ³ -atm/kmole/K
r_{FT}	reaction rate for FT synthesis, kmol/kg-cat.s

r_k	reaction rate for kth reaction, kmol/kg-cat.s
r_{WGS}	reaction rate for water gas shift, kmol/kg-cat.s
T	Reaction temperature [K]
T_c	Critical temperature [K]
T_{ri}	Reduced temperature of component i
T_w	reactor wall temperature, K
U_G	gas superficial velocity, m/s
U_{G0}	inlet gas superficial velocity, m/s
U_L	liquid superficial velocity, m/s
U_s	settling velocity of catalyst particles in swarm, m/s
U_t	terminal settling velocity of a single particle, m/s
v	dimensionless gas-phase superficial velocity (U_G/U_{G0})
V	Moles of vapor in flash calculations
V_s	solvent molar volume ($m^3/kmol$)
V_m	theoretical close-packed volume for solvent spheres($10^{-6}m^3/mol$)
w	dimensionless catalyst concentration
x_i	dimensionless liquid-phase concentration of component i ($C_{L,i}H_i/P$) in slurry reactor model or mole fraction of component i in liquid phase in flash calculations
y_i	mole fraction of component i in the vapor (or gas) phase
z	dimensionless axial distance (x/L)
z_i	Overall Mole fraction of component i in feed in flash calculations
Z	Compressibility factor of the liquid or vapor phase

Dimensionless Numbers

- Ar : Archimedes number $[\rho_L(\rho_{cat}-\rho_L)d_p^3/\mu_L^2]$
- Bo : Bond number $(D_c^2\rho_{sl}g/\sigma_L)$
- Fr_G : Froude number for gas (U_G^2/gD_c)
- Pe_G : gas-phase Peclet number $(U_G L/D_G\epsilon_G)$
- Pe_L : liquid-phase Peclet number $(U_L L/D_L\epsilon_L)$
- Pe_S : solid-phase Peclet number $[(U_S - U_L/\epsilon_L)(L/D_S)]$
- Re_G : gas Reynolds number $(U_G D_c \rho_L/\mu_L)$
- Re_p : particle Reynolds number $(U_L d_p \rho_L/\mu_L)$
- St_G : gas-phase Stanton number $(K_{L,i} a L/U_{G0})$
- $St_{L,i}$: liquid-phase Stanton number for component i $(K_{L,i} a L/U_L)$

Greek Letters

- δ_{ij} Binary interaction parameter in Peng-Robinson EOS with one-parameter mixing rule
- Φ_i^V Fugacity coefficient of component i in vapor phase [atm]
- Φ_i^L Fugacity coefficient of component i in liquid phase [atm]
- ω Accentric factor
- θ_{ik} stoichiometric coefficient of component i in reaction k
- ϵ_G gas holdup
- ϵ_L liquid holdup
- $\epsilon_{s,0}$ volume fraction of solids at reactor bottom
- $\epsilon_{s,ave}$ average volume fraction of solids in reactor

ρ_L	density of the liquid phase, kg/m^3
ρ_{SL}	density of slurry, kg/m^3
ρ_w	density of water, kg/m^3
σ_L	surface tension of the hydrocarbon solvent, N/m
σ_w	surface tension of water, N/m
α	probability of chain growth
β	ratio of hydrostatic head to the head pressure
μ_L	viscosity of hydrocarbon solvent, $\text{Pa}\cdot\text{s}$
μ_w	viscosity of water, $\text{Pa}\cdot\text{s}$
γ_i	stoichiometric coefficient for component i in FT synthesis reaction, ($i = \text{CO}, \text{H}_2, \text{H}_2\text{O}, \text{HC}$)
ξ_i	molecular diameter of component i (\AA)
ξ_s	molecular diameter of solvent molecules (\AA)

Subscripts

L	liquid phase
G	gas phase
i	component i
k	k th reaction
o	inlet condition
S	solid phase

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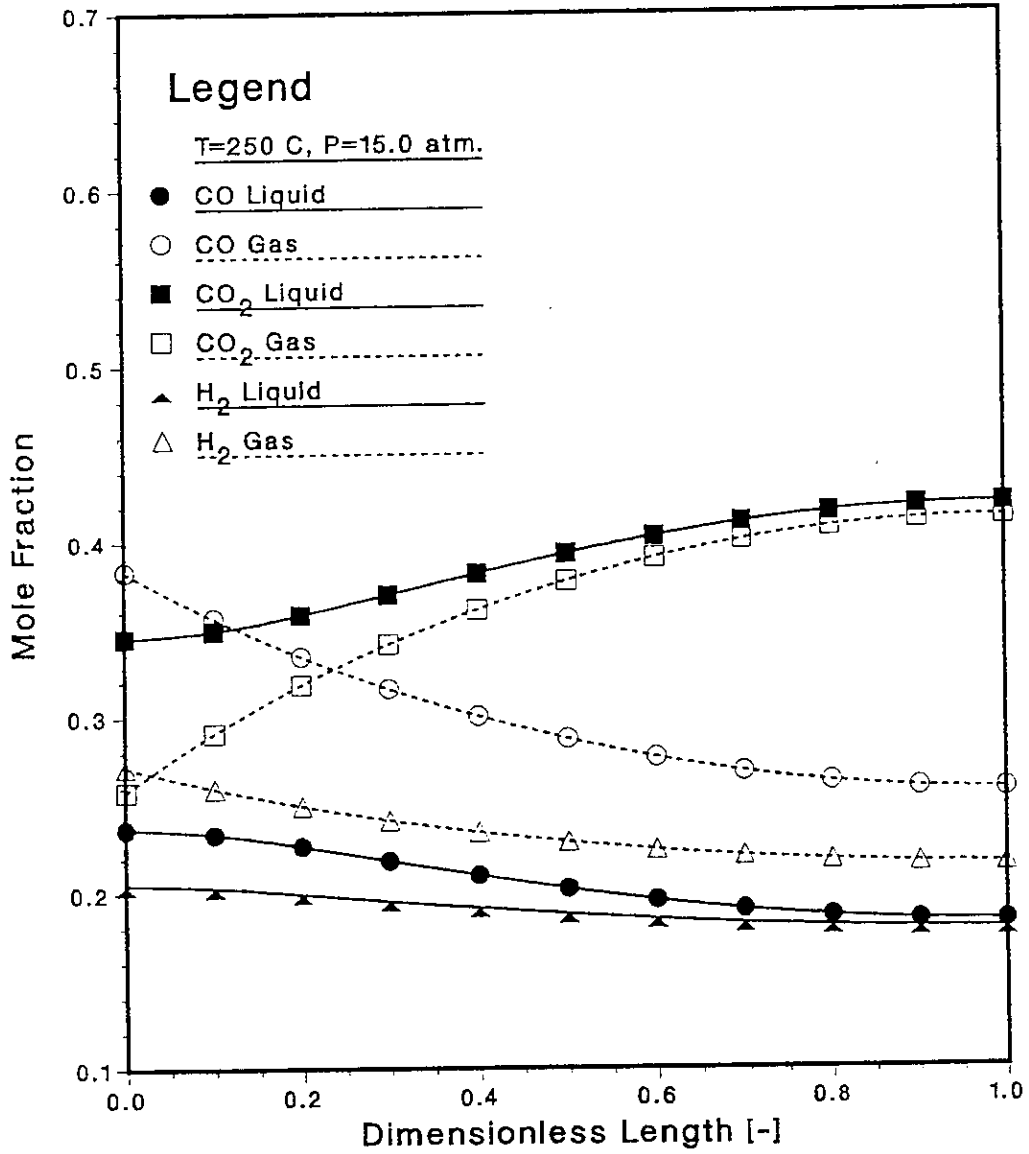


Figure 1: Concentration Profiles in Bubble Column Slurry Reactor

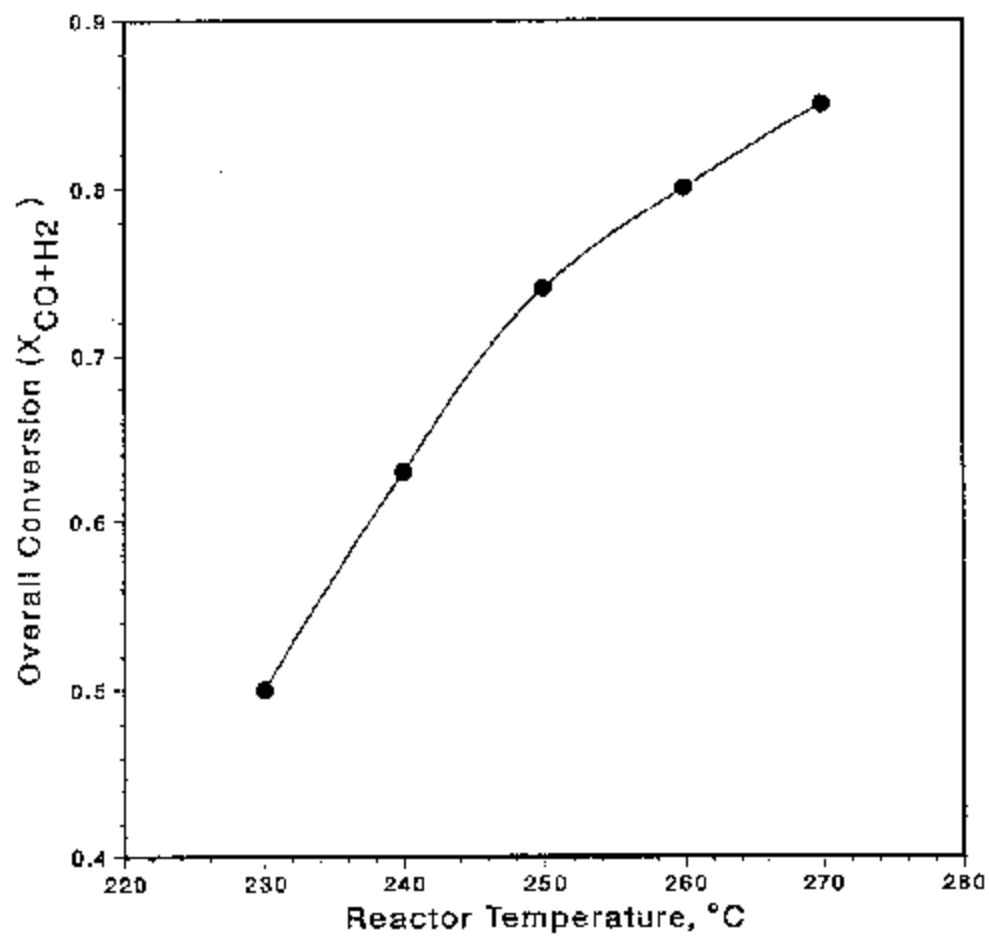


Figure 2: Effect of Reactor Temperature on Syn Gas Conversion

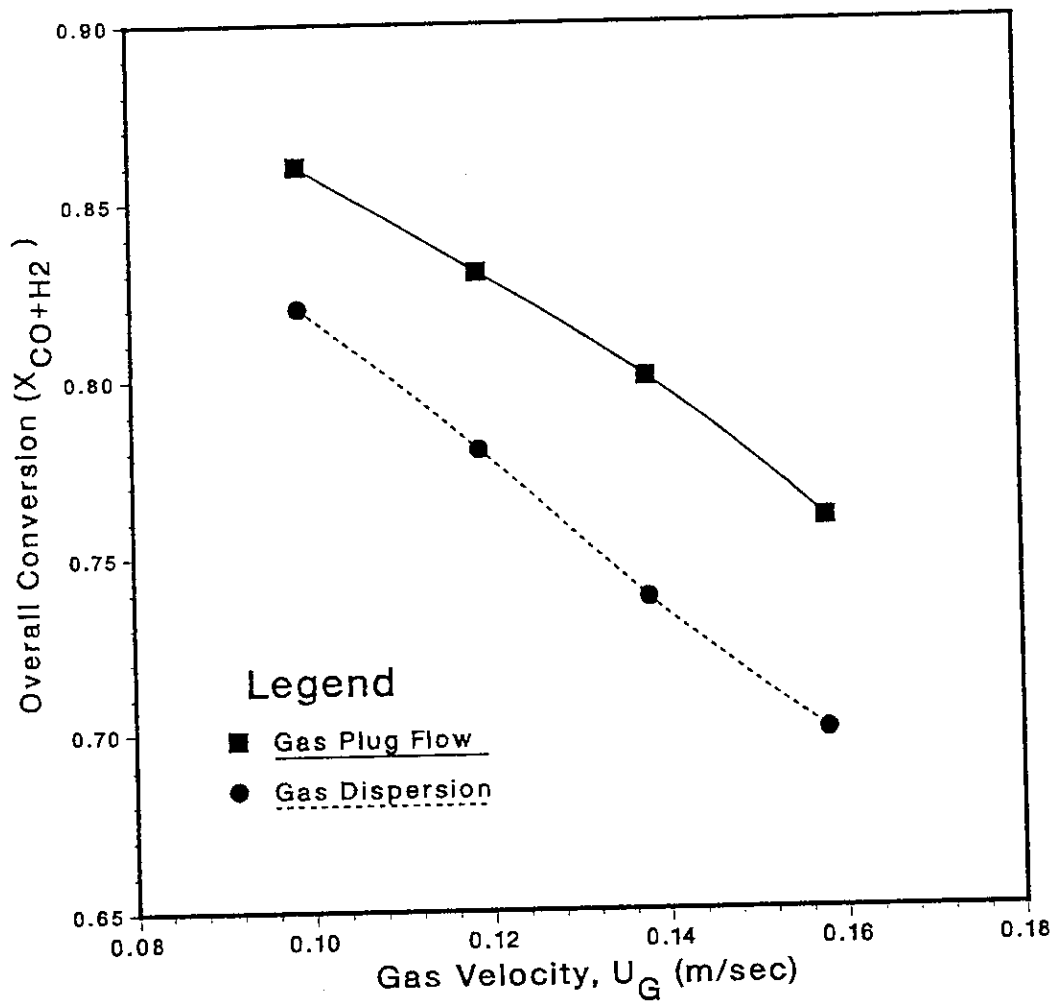


Figure 3: Effect of Gas Velocity on Syn Gas Conversion

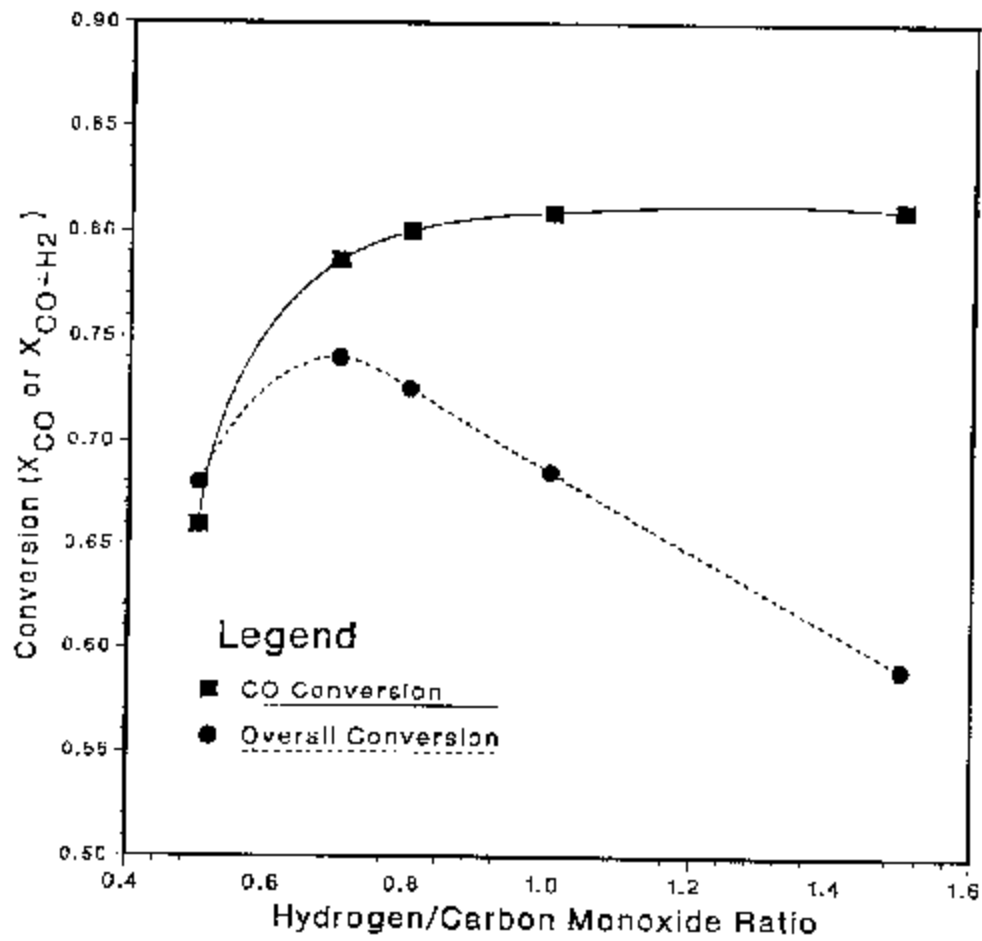


Figure 4: Effect of Hydrogen/Carbon Monoxide Ratio on Syn Gas Conversion

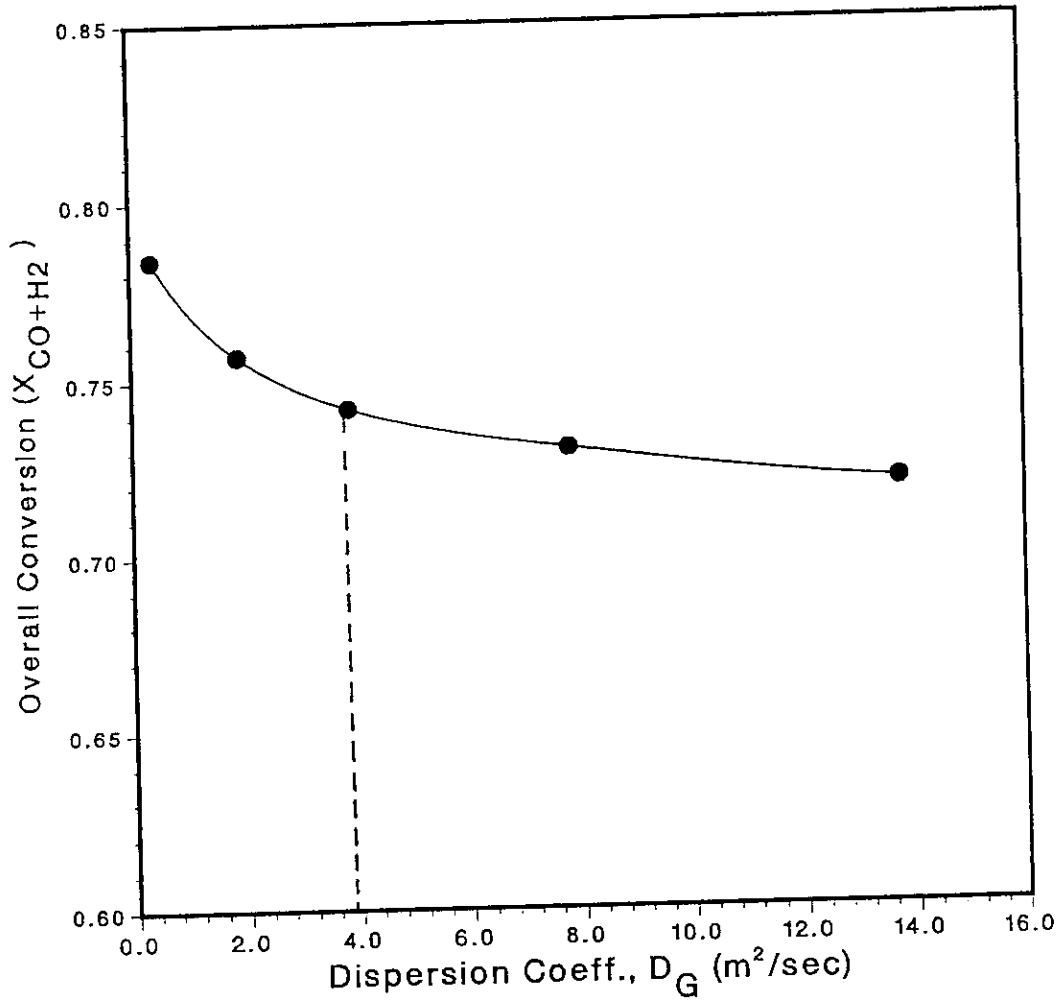


Figure 5: Effect of Gas Phase Dispersion Coefficient on Syn Gas Conversion

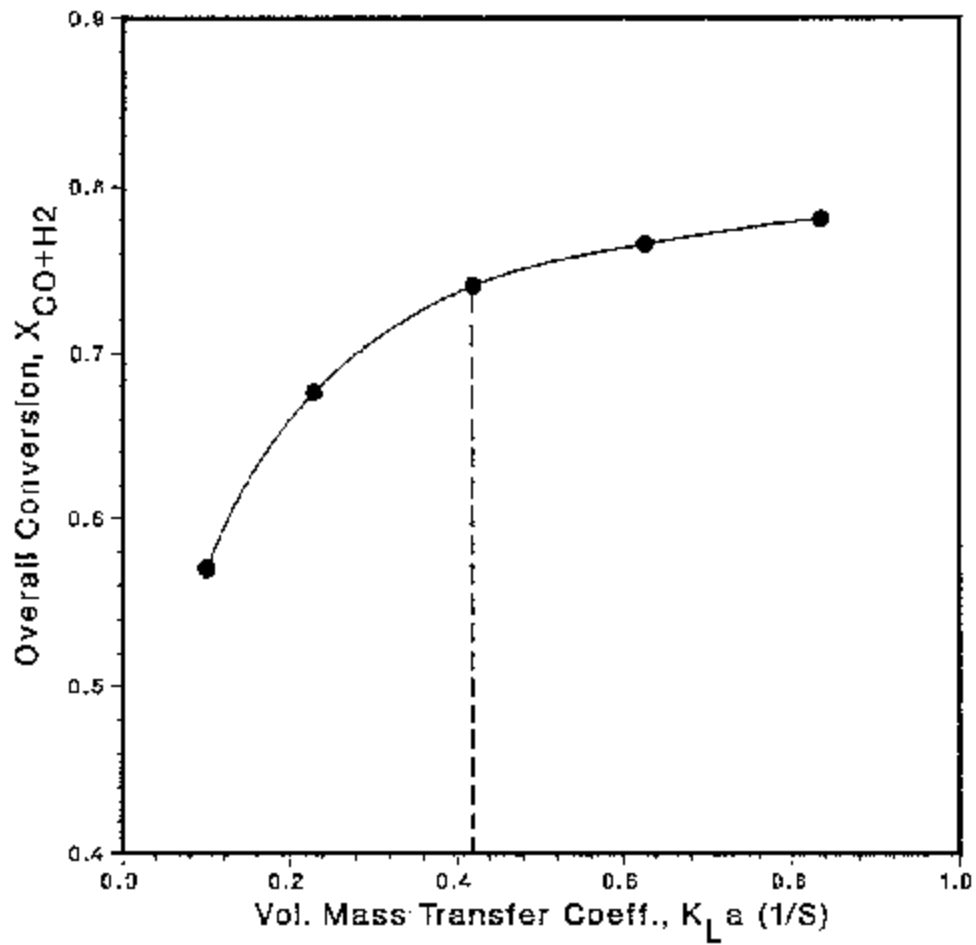


Figure 6: Effect of Volumetric Mass Transfer Coefficient on Syn Gas Conversion