4.0 Recommendations for Future Work:

Hydrodynamics of slurry bubble columns need further investigations especially the effects of high solids concentrations (>30.0 wt%) and high pressure on gas holdup need to be investigated. At increasing solids concentrations in a slurry reactor, there is a possibility of poor gas-liquid contact especially in the distributor region. This problem was pointed out during our analysis of Air Products PDU data for methanol synthesis. Therefore, new studies for hydrodynamics in slurry bubble columns should also investigate the effects of solid concentration and gas velocity on mixing patterns and gas-slurry contact. This brings us to the problem of gas-liquid mass transfer in slurry bubble columns. Poor gas-liquid contact would result in low gas-liquid mass transfer and could lower the reactor productivity. Gas holdup and volumetric mass transfer coefficient are interrelated via the interfacial area. It would be, therefore, prudent to conduct hydrodynamic and mass transfer studies simultaneously.

Future hydrodynamic studies in slurry bubble columns should also include the effects of decreasing gas velocity and presence of internals. Superficial gas velocity in a Fischer-Tropsch reactor reduces to about half its initial value from bottom to the reactor top. Moreover, internal heat exchanger in a Fischer-Tropsch reactor could occupy up to 15% of reactor cross-sectional area. Little work has been done so far to investigate the effect of decreasing gas velocity and presence of internals on hydrodynamic parameters such as gas holdup, gas and slurry phase mixing and axial solids distribution. These studies should be carried out in large diameter reactors (>0.2 m) and should also include regime mapping and investigate heterogeneities in slurry bubble columns.

Future studies for hydrodynamics and gas-liquid mass transfer in slurry bubble columns should be conducted using high molecular weight Fischer-Tropsch waxes (C_{30}-C_{50}) or other high molecular weight hydrocarbon solvents with similar properties. As pointed out in this report, gas-liquid mass transfer studies in slurry bubble columns, so far, have been conducted using aqueous solutions at ambient temperatures.

The scope of reactor models developed in this study could be further improved when more information for hydrodynamics and mass transfer become available.

In our reactor model we used Henry's law to determine gas solubilities at reactor conditions. Gas solubilities could be more accurately determined by using a suitable equation of state (i.e Peng Robinson). Reactor model can be modified to include such calculations.

Rate constants in a kinetic expression vary with catalyst activity. Catalyst activity in turn varies widely depending on the procedures used for catalyst preparation and activation. Mechanistic kinetic models are expected to provide better predictions of reaction rates over wide range of operating conditions. Our analysis of kinetic models for methanol synthesis showed that reliable mechanistic model for methanol synthesis still needs to be developed.

The FT synthesis reactor model can be further tested and refined as more data become available from demonstration units.

The reactor models developed in this study can be easily integrated with a process simulator to improve overall process design and optimization.

5.0 Conceptual Commercial size Reactor Design:

Using the computer models developed in this study, commercial size slurry reactors for methanol synthesis and Fischer-Tropsch synthesis can be sized rather quickly. Appendix-B and C contain sample input and output files for Methanol synthesis and Fischer-Tropsch synthesis models respectively. The reactor models can also be used to obtain optimum reactor size for a required rate of production. For this report, we sized a methanol synthesis reactor for a production rate of 1000 metric ton per day and a Fischer-Tropsch synthesis reactor for production rate of 400 metric tons per day of C_3^+ products. For commercial size slurry reactors, Bechtel has recommended an upper limit of 4.8 m for reactor diameter (Fox and Degen, 1990). The operating conditions for commercial size reactors were selected based on literature review and our analysis of process development units data. Table 9 and 10 summarize the dimensions and model outputs for methanol synthesis and Fischer-Tropsch synthesis reactors respectively.

Table 9. Commercial size reactor for Methanol Synthesis

Diameter	4.8 m
Length	12.5 m
Temperature	240°C
Pressure	55.0 atm
Slurry Conc.	34 wt%
Gas velocity	0.155 m/s
H_2/CO ratio	0.64
Catalyst size	0.000025 m
Tube OD	0.038 m
Tube ID	0.034 m
Tube Length	12.0 m
Total Cooling Surface	1134.0 m^2
No. of tubes	800
Methanol Production	1000.0 MTD
STY	188.0 kg methanol/hr.m^3
Productivity	0.745 kg methanol/hr-kgcat
Space Velocity	1340.0 $Nm^3/hr\text{-}m^3$
Gas Holdup	0.30

Table 10. Commercial size reactor for Fischer-Tropsch synthesis

Diameter	4.8 m
Length	11.5 m
Temperature	255°C
Pressure	28.0 atm
Slurry Conc.	34 wt%
Gas velocity	0.155 m/s
H_2/CO ratio	0.64
Catalyst size	0.000025 m
Tube OD	0.038 m
Tube ID	0.034 m
Tube Length	11.0 m
Total Cooling Surface	3400 m^2
No. of tubes	2600
C_3^+ Production	411.0 MTD
STY	100.0 kg HC/hr.m^3
Productivity	0.385 kg HC/hr-kgcat
Space Velocity	640.0 $Nm^3/hr\text{-}m^3$
Gas Holdup	0.272

5.1 Cost Comparison with Fixed Bed Reactor:

Design information for fixed-bed reactors was mainly obtained from literature although some quick checks were made using the heterogeneous reactor model for fixed-bed reactor. Fixed bed reactor design for methanol synthesis is well established. The commonly used fixed-bed reactors for methanol synthesis are ICI quench type reactor and Lurgi shell and tube type reactor. The Lurgi reactor design offers advantages over ICI design. The overall dimensions of a commercial size Lurgi fixed-bed reactor for a production rate of 1480 MTD are provided in the Bechtel report (Fox and Degen, 1990). The conventional fixed bed reactors for methanol synthesis use stoichiometric synthesis gas. In order to make a one-for-one comparison of reactor design, a slurry reactor for methanol synthesis was sized using stoichiometric synthesis gas (see Table 11). Figure 17 and 18 show the dimensions of fixed bed and slurry reactors respectively for methanol synthesis. The length of slurry reactor required for similar production rate is about twice that of the fixed-bed reactor. This difference is mainly due to significantly higher catalyst loading possible in a fixed bed reactor. The conventional fixed bed reactor, however, is not suitable to process syngas from with low hydrogen to carbon monoxide ratio due to higher rate of deactivation and possibility of plugging from coke formation. The operating cost of a fixed bed reactor is expected to be higher due to frequent shut down for catalyst regeneration.

For Fischer-Tropsch synthesis tubular fixed-bed ARGE reactors have been used at Sasol, South Africa. Bechtel used the design basis for ARGE reactors (information given in Encyclopedia of Chemical Technology, 2nd Edition, Vol. 4) to size a larger tubular fixed bed reactor for a production capacity of 400 MTD. Again for cost comparisons, slurry reactor for Fischer-Tropsch synthesis was sized for similar production rate, using our reactor model (Table 12). The dimensions of slurry and fixed-bed reactors are shown in Figures 19 and 20 respectively.

In order to obtain realistic cost estimates, several well known manufacturers of large size high pressure equipments were contacted. Most of them, however, expressed their inability to fabricate vessels of these sizes. We are presently in touch with one company willing to provide the cost estimates for these reactors.

Table 11. Slurry reactor design for methanol synthesis for cost comparison

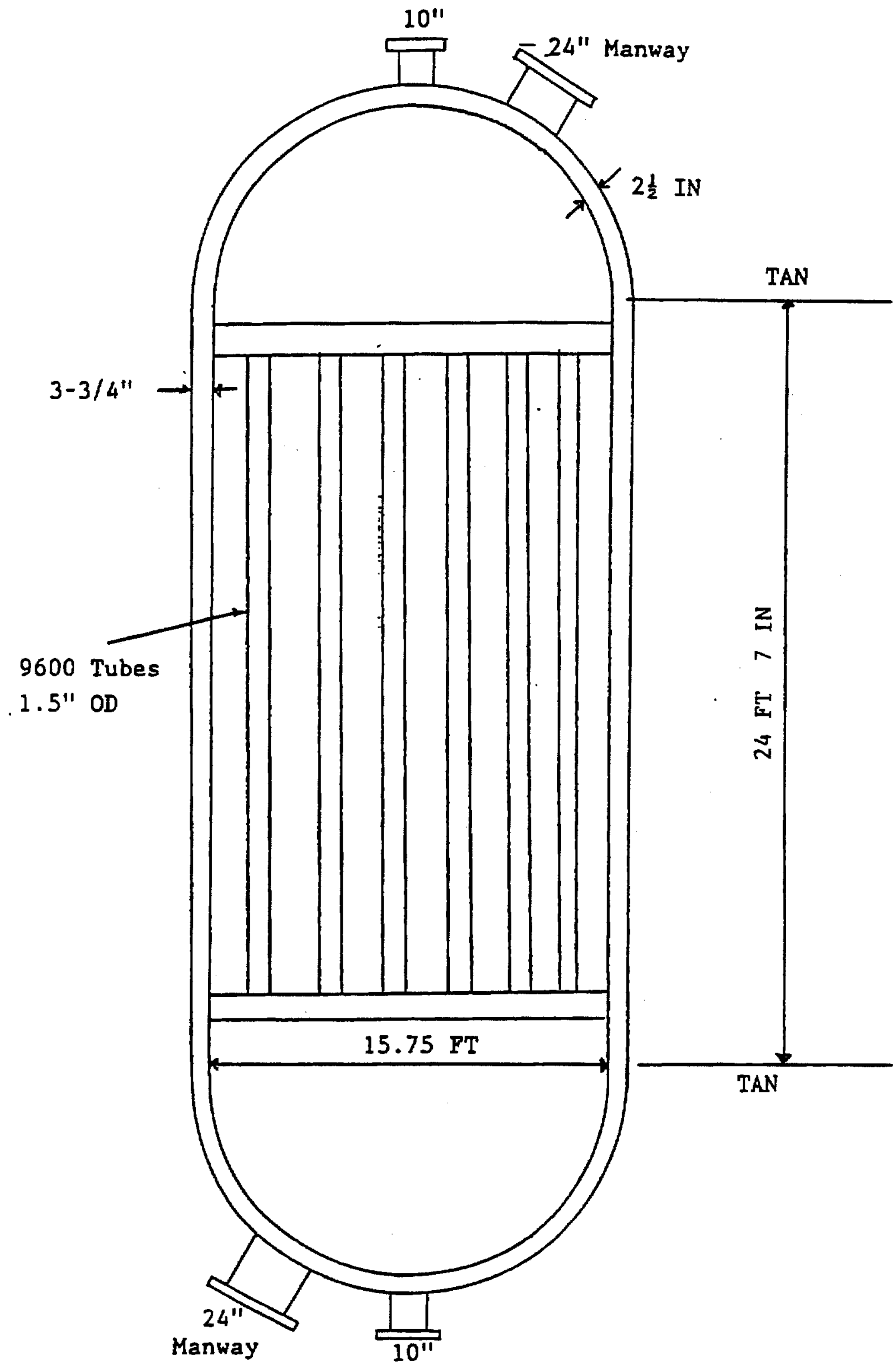
Diameter	4.8 m
Length	13.5 m
Temperature	240°C
Pressure	55.0 atm
Slurry Conc.	34 wt%
Gas velocity	0.155 m/s
$(H_2-CO_2)/(CO+CO_2)$	2.05
Catalyst size	0.000025 m
Tube OD	0.038 m
Tube ID	0.031 m
Tube Length	13.0 m
Total Cooling Surface	2100.0 m^2
No. of tubes	1500
Methanol Production	1480.0 MTD
STY	266.0 kg methanol/hr.m^3
Productivity	1.0 kg methanol/hr-kgcat
Space Velocity	1170.0 $Nm^3/hr-m^3$
Gas Holdup	0.30

Table 12. Slurry reactor design for FT synthesis for cost comparison

Diameter	4.8 m
Length	11.5 m
Temperature	255°C
Pressure	28.0 atm
Slurry Conc.	34 wt%
Gas velocity	0.155 m/s
H_2/CO ratio	0.64
Catalyst size	0.000025 m
Tube OD	0.038 m
Tube ID	0.034 m
Tube Length	11.0 m
Total Cooling Surface	3300 m^2
No. of tubes	2500
C_3^+ Production	411.0 MTD
STY	100.0 kg HC/hr.m^3
Productivity	0.385 kg HC/hr-kgcat
Space Velocity	640.0 $Nm^3/hr\text{-}m^3$
Gas Holdup	0.27

Figure 17

Fixed BED Methanol Reactor
(Lurgi Design)

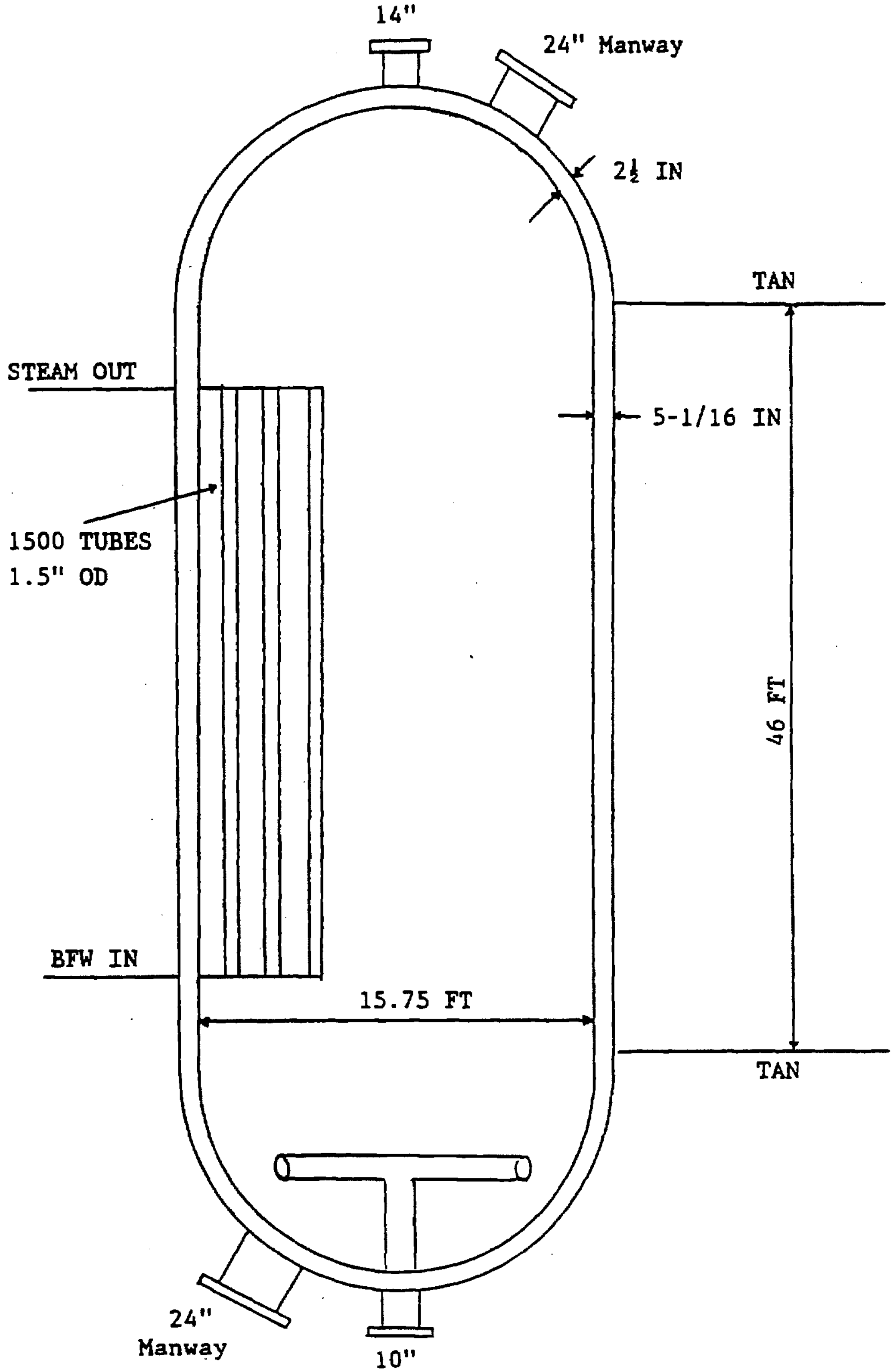


Design Conditions - Tube Side 875 PSIG 550F; Shell Side 650 PSIG 550F

Metallurgy: Shell - SA516 GR70 with 1/8" C.A.; Tubes C.S. Chromized

Figure 18

Slurry Reactor for Methanol Synthesis

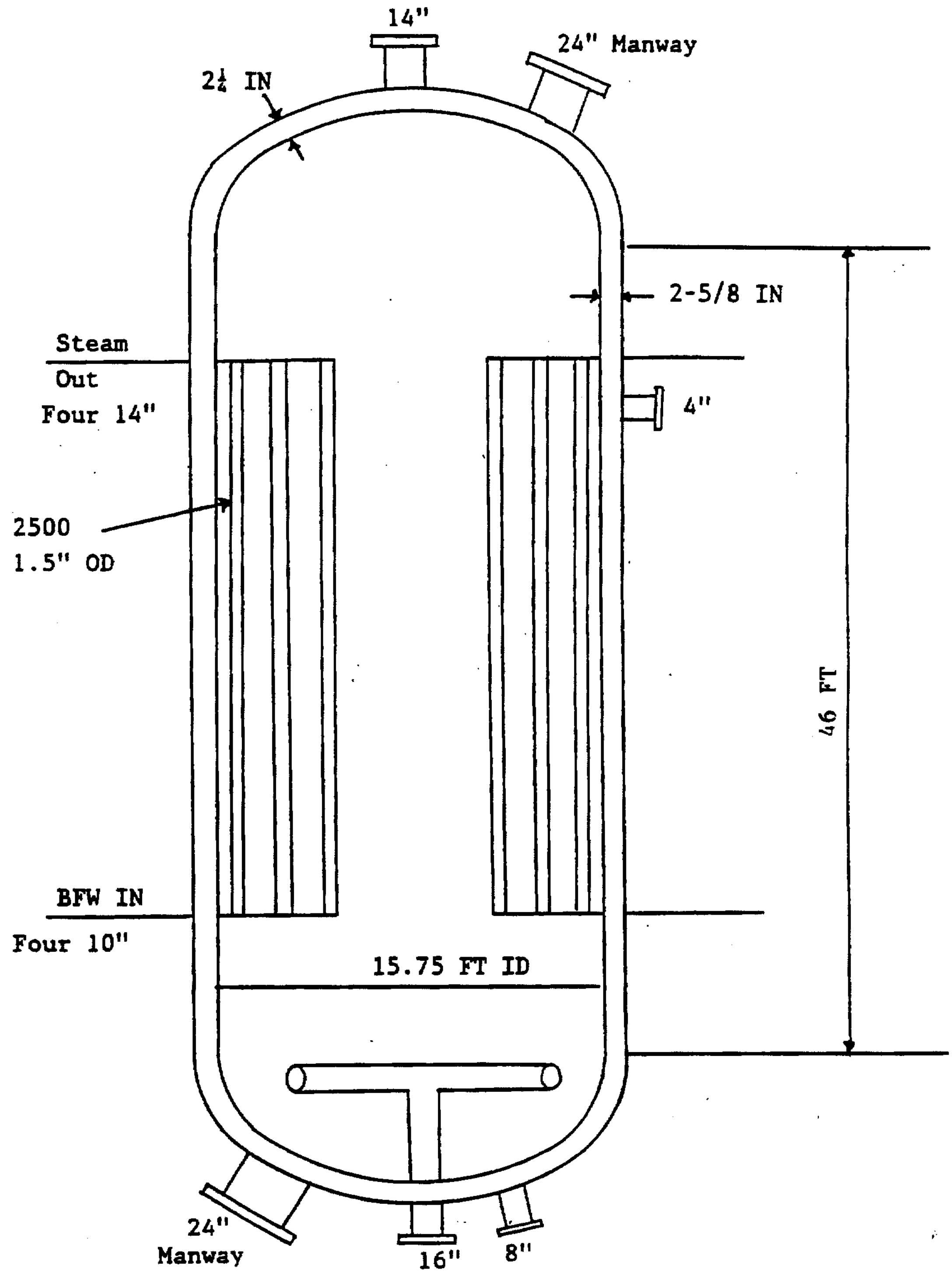


Design Conditions - Tube Side 580 PSIG 550F; Shell Side 875 PSIG 550F

Metallurgy: Shell - SA516 GR70 with SS304 Cladding Tubes SS304

Figure 19

Slurry Reactor Fischer - Tropsch Synthesis

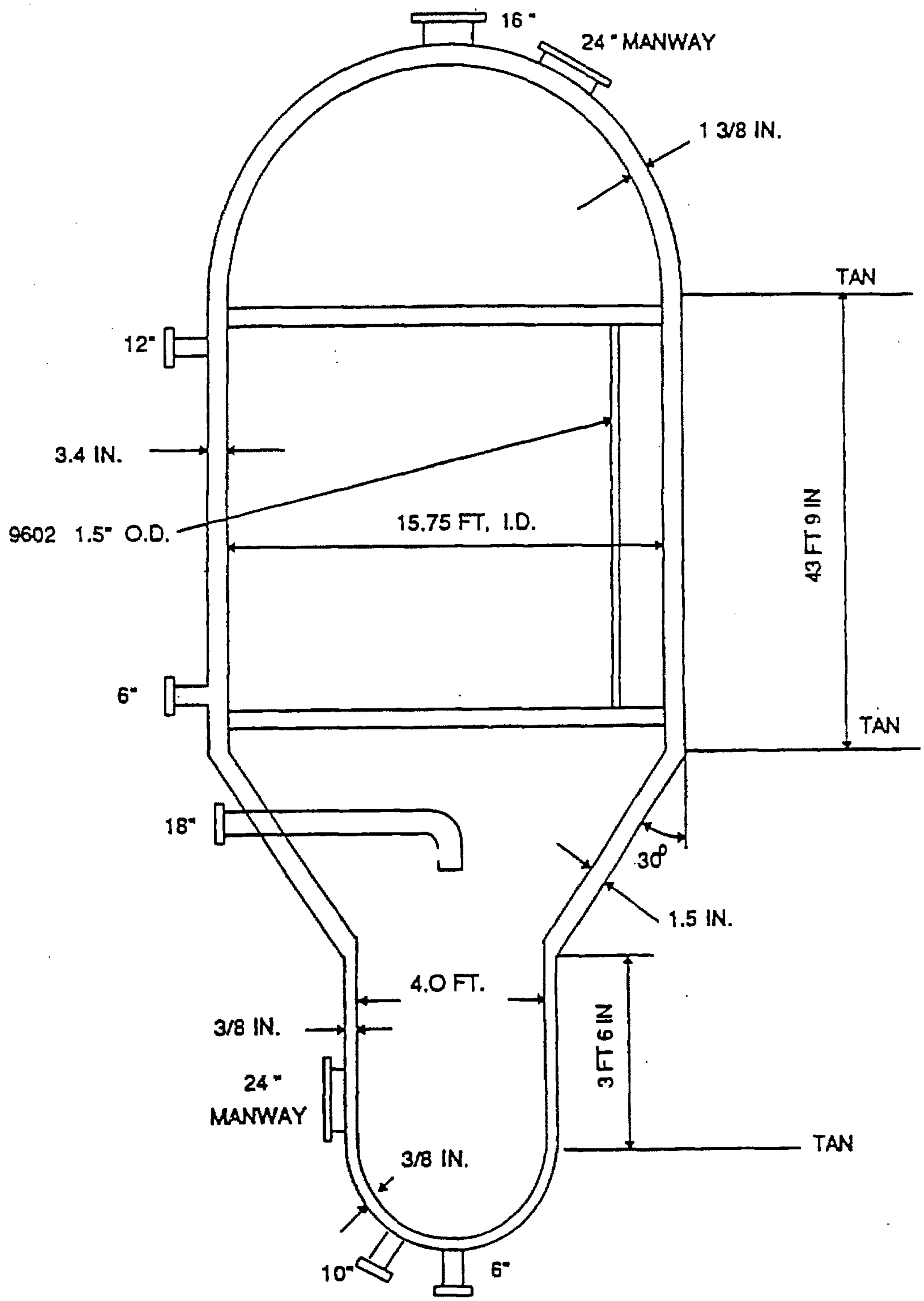


Design Conditions - Tube Side-580 PSIG 550F; Shell Side 460 PSIG 550F

Metallurgy: Shell - SA516 GR70 with 1/8" C.A.; Tubes C.S. Chromized

Figure 20

Fixed Bed Fischer - Tropsch Reactor
(Bechtel Design)



Design Conditions - Tube Side: 460 PSIG 550F; Shell Side: 580 PSIG 550F

Metallurgy: Shell - SA516 GR70 with 1/8" C.A.; Tubes C.S. Chromized

NOMENCLATURE

a_1	water adsorption coefficient FT synthesis, Equations (65),(68)
a_{mix}	Peng-Robinson attractive parameter for mixture [atm cm^6 / $gmole^2$]
a_i,a_j	Peng-Robinson attractive parameter for component i and j [atm cm^6 / $gmole^2$]
A_i,B_i	coefficient and exponent for equation of Henry's constant
b_1	water adsorption coefficient FT synthesis, Equation (67), 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 27, a function of molecular size of solute and solvent
C_a, C_b	correction factors in the Peng-Robinson EOS
C_{cat}	catalyst concentration, kg/m^3 slurry
$C_{cat,avg}$	average catalyst concentration, kg/m^3 slurry
$C_{cat,b}$	catalyst concentration at reactor bottom, kg/m^3
$C_{G,i}$	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_p	particle diameter, m
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_o	ratio of hydrostatic head to the head pressure
H_i	Henry's constant for component i, $atm.m^3/kmol$
k_o	rate constant for FT synthesis rate by Equation (65), kmol/kg-cat.s.atm
k_1,k_2	reaction rate constants (units depend on kinetic model)
k_c	rate constant for FT synthesis on cobalt catalyst kmol/kg-cat.s.atm
$k_{L,i}$	liquid-side mass transfer coefficient for component i, (m/s)
$k_{ps,1},k_{ps,2},k_{ps,3}$	pseudo reaction rate constants, equations (43)to(45)
k_{wo}	rate constant for water gas shift reaction by Equation (68), kmol/kg-cat.s.atm
K_{c1},K_{c2},K_{c3}	chemical equilibrium constants based on concentration
K_P	chemical equilibrium constants based on partial pressure
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/kmol)
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 ($i=CO,H_2,CO_2,H_2O$)
P_{ri}	reduced pressure of component i
P_T	pressure at reactor top, atm
R	universal gas constant, 0.082 m^3-atm/kmole/K
r_{MeOH}	reaction rate for MT synthesis, kmol/kg-cat.s
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_{Go}	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_{GO})
V	specific volume of gas in equation of state or moles of vapor in flash calculations
V_s	solvent molar volume (m^3/kmol)

V_{so} theoretical close-packed volume for solvent spheres($10^{-6}m^3/mol$)

w dimensionless catalyst concentration ($C_{cat}/C_{cat,avg}$)

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 $(U_{Go}L/D_G\epsilon_G)$

Pe_L : liquid-phase Peclet number $(U_LL/D_L\epsilon_L)$

Pe_P : particle peclet number (U_GD_c/D_s)

Pe_S : solid-phase Peclet number $[(U_S - U_L/\epsilon_L)(L/D_S)$

Re_G : gas Reynolds number $(U_GD_C\rho_L/\mu_L)$

Re_p : particle Reynolds number $(U_td_p\rho_L/\mu_L)$

St_G : gas-phase Stanton number $(K_{L,i}aL/U_{Go})$

$St_{L,i}$: liquid-phase Stanton number for component i $(K_{L,i}aL/U_L)$

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
ϵ_s	solids holdup
$\epsilon_{s,o}$	volume fraction of solids at reactor bottom
$\epsilon_{s,sus}$	average volume fraction of solids in slurry
ρ	density, 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	fraction of type I site on FT catalyst
μ	viscosity, Pa.s

μ_w viscosity of water, Pa.s

ν kinematic viscosity, m^2/s

γ_i stoichiometric coefficient for component i in FT synthesis reaction,($i=CO,H_2,H_2O,HC$)

λ fraction of paraffinic hydrocarbons

ξ_i molecular diameter of component i (A)

ξ_s molecular diameter of solvent molecules (A)

<u>Subscripts</u>

L liquid phase

G gas phase

i component i

k kth reaction

o inlet condition

S solid phase

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