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A novel slurry bubble column membrane reactor concept for Fischer–Tropsch synthesis in GTL technology

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ABSTRACT

Fischer–Tropsch synthesis (FTS) plays an important role in the production of ultra-clean transportation fuels, chemicals, and other hydrocarbon products. In this work, a novel combination of fixed-bed and slurry bubble column membrane reactor for Fischer–Tropsch synthesis has been proposed. In the first catalyst bed, the synthesis gas is partially converted to hydrocarbons in a water-cooled reactor which is fixed bed. In the second bed which is a membrane assisted slurry bubble column reactor, the heat of reaction is used to preheat the feed synthesis gas to the first reactor. Due to the decrease of H_2/CO to values far from optimum reactants ratio, the membrane concept is suggested to control hydrogen addition. A one-dimensional packed-bed model has been used for modeling of fixed-bed reactor. Also a one-dimensional model with plug flow pattern for gas phase and an axial dispersion pattern for liquid-solid suspension have been developed for modeling of slurry bubble column reactor. Proficiency of a membrane FTS reactor (MR) and a conventional FTS reactor (CR) at identical process conditions has been used as a basis for comparison in terms of temperature, gasoline yield, H_2 and CO conversion as well as selectivity. Results show a favorable temperature profile along the proposed concept, an enhancement in the gasoline yield and, thus a main decrease in undesirable product formation. The results suggest that utilizing this type of reactor could be feasible and beneficial. Experimental proof of concept is needed to establish the validity and safe operation of the proposed reactor.

Keywords: Bubble column; Slurry reactor; Fischer–Tropsch synthesis; Hydrogen-permselective membrane; GTL technology

1. Introduction

1.1. *GTL technology*

Recently, the high oil price has created considerable interest in the development of alternative technology for the manufacture of transportation fuels. Natural gas in remote areas can be converted to shippable liquid fuels (e.g., gasoline, diesel, and wax) through the gas-to-liquid (GTL) process (Schulz, 1999; Hall, 2005). In the GTL process, Fischer–Tropsch synthesis (FTS) is the key technology for converting synthesis gas (mixture of CO and H_2) to liquid fuels. The development of an effective catalyst and reactor system is the most competitive issue in FTS. Owing to the high demand on gasoline in the world and its higher price relative to that of diesel, production of gasoline from the FT process becomes more favorable. FTS is either low temperature process (LTFT) or high temperature process (HTFT) depending on the product required. High temperature process operates at 300–350 °C on Fe-based catalysts and is mainly used for the production of $C5^+$ and linear olefins while the low temperature process operates at 200–240 °C and is applied for the production of waxy material (Akhtar et al., 2006). The various types of reactors (including fixed-bed, fluidized bed, and slurry phase) have been considered in the history of FTS process development, characterizing with the most suitable particle size of the catalyst used (Wang et al., 2003).

Abbreviations: CR, conventional reactor; FMDR, fluidized-bed membrane dual type reactor; FTS, Fischer–Tropsch synthesis; HTFT, high temperature Fischer–Tropsch process; LTFT, low temperature Fischer–Tropsch process.

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Nomenclature

A_c cross section area of each tube, m^2
A_i inner area of each tube, m^2
A_o outer area of each tube, m^2
A_s lateral area of each tube, m^2
A_{shell} cross section area of shell, m^2
a interface area between gas and slurry phase, m^2
a_v specific surface area of catalyst pellet, $m^2\,m^{-3}$
a_w cooling tube specific external surface area referred to the total reactor volume, $m^2\,m^{-3}$
$c_{i,g}$ concentration of i in the gas phase at reactor inlet, $mol\,m^{-3}$
$c_{i,l}$ concentration of i in the liquid phase at reactor inlet, $mol\,m^{-3}$
c_{Pg} specific heat of the gas at constant pressure, $J\,mol^{-1}\,k^{-1}$
c_{ph} specific heat of the hydrogen at constant pressure, $J\,mol^{-1}\,k^{-1}$
c_{Ps} specific heat of the catalyst at constant pressure, $J\,mol^{-1}\,k^{-1}$
c_{psl} heat capacity of the liquid–solid suspension, $J\,mol^{-1}\,k^{-1}$
C_s solid volume fraction in gas free slurry, $kg\,m^{-3}$
c_s solid hold up in liquid phase, $g\,m^{-3}$
c_t total concentration, $mol\,m^{-3}$
D_i tube inside diameter, m
D_{ij} binary diffusion coefficient of component i in component j, $m^2\,s^{-1}$
$D_{i,m}$ diffusion coefficient of component i in the mixture, $m^2\,s^{-1}$
D_o tube outside diameter, m
DT Reactor diameter, m
d_p particle diameter, m
E_g axial dispersion coefficient of gas phase, $m^2\,s^{-1}$
E_l axial dispersion coefficient of liquid phase, $m^2\,s^{-1}$
F_t total molar flow rate in shell side, $mol\,s^{-1}$
F^{sh} total molar flow in shell side, $mole\,s^{-1}$
F^t total molar flow per tube, $mole\,s^{-1}$
F^l molar flow in liquid side, $mole\,s^{-1}$
F^g molar flow in gas side, $mole\,s^{-1}$
F_{t0} total molar flow rate in tube side, $mol\,s^{-1}$
g accelerating due to gravity, $m\,s^{-2}$
H Henry's coefficient, Pa
h_f gas–solid heat transfer coefficient, $W\,m^{-2}\,K^{-1}$
h_i heat transfer coefficient between the gas phase in the tube side and reactor wall, $W\,m^{-2}\,K^{-1}$
h_o heat transfer coefficient of boiling water in the shell side, $W\,m^{-2}\,K^{-1}$
K conductivity of fluid phase, $W\,m^{-1}\,K^{-1}$
K_{bei} mass transfer coefficient for component i in fluidized-bed, $m\,s^{-1}$
K_w thermal conductivity of reactor wall, $W\,m^{-1}\,K^{-1}$
k_{gi} mass transfer coefficient of component i in gas-phase, $m\,s^{-1}$
L length of reactor, m
M_i molecular weight of component i, $g\,mol^{-1}$
N number of components
P_H^t hydrogen partial pressure in tube side, bar
P_H^{sh} hydrogen partial pressure in shell side, bar
$\bar{P}$ permeability of hydrogen through Pd–Ag layer, $mol\,m^{-1}\,s^{-1}\,Pa^{-1/2}$
P_0 pre-exponential factor of hydrogen permeability, $mol\,m^{-1}\,s^{-1}\,Pa^{-1}$
P_{sv} saturated vapor pressure, Pa
$\Delta P/\Delta z$ pressure gradient along the column wall, $Pa\,m^{-1}$
R universal gas constant, $J\,mol^{-1}\,K^{-1}$
Re Reynolds number
R_i inner radius of Pd–Ag layer, m
R_o outer radius of Pd–Ag layer, m
R_p particle diameter, m
r_i reaction rate of component i, $mol\,kg^{-1}\,s^{-1}$
Sc_i Schmidt number of component i
T bulk gas phase temperature, K
T_c cooling temperature, K
T_s temperature of solid phase, K
T_{shell} temperature of coolant stream, in first reactor, K
T_t temperature of synthesis gas in tube side, K
U_{shell} overall heat transfer coefficient between coolant and process streams (gas-cooled reactor), $W\,m^{-2}\,K^{-1}$
U_{tube} overall heat transfer coefficient between coolant and process streams (water-cooled reactor), $W\,m^{-2}\,K^{-1}$
U_{df} superficial velocity of gas through the dense phase, $m\,s^{-1}$
u superficial velocity of liquid phase, $m\,s^{-1}$
u_g superficial gas velocity, $m\,s^{-1}$
u_l linear velocity of liquid phase, $m\,s^{-1}$
u_{sg} superficial gas velocity in slurry reactor, $m\,s^{-1}$
u_{ss} superficial slurry velocity, $m\,s^{-1}$
y_i mole fraction of component i in the fluid phase, $mol\,mol^{-1}$
y_{is} mole fraction of component i in the solid phase, $mol\,mol^{-1}$
y_{ig} mole fraction of component i in the gas phase, $mol\,mol^{-1}$
y_{il} mole fraction of component i in the liquid phase, $mol\,mol^{-1}$
w_c weight fraction catalyst in gas free slurry
z axial reactor coordinate, m

Greek letters

α_{eff} slurry to internal coil wall conversion heat transfer coefficient, $W\,m^{-2}\,K^{-1}$
α_H hydrogen permeation rate constant, $mol\,m^{-1}\,s^{-1}\,Pa^{-0.5}$
$\Delta H_{f,i}$ enthalpy of formation of component i, $J\,mol^{-1}$
ΔP pressure difference, kPa
Δz a small increment in vertical distance, m
ε_B void fraction of catalyst bed
ε_g gas holdup
ε_l liquid holdup
ε_s void fraction of catalyst
ρ density of fluid phase, $kg\,m^{-3}$
ρ_B density of catalytic bed, $kg\,m^{-3}$
ρ_g density of gas phase, $kg\,m^{-3}$
ρ_l density of liquid phase, $kg\,m^{-3}$