6. CONCLUSIONS

This work has focused on pilot reactor studies and modeling of the wall–cooled fixed bed reactor for highly exothermic reactions. Special attention has been laid on the subject of heat transfer in such systems because this is of vital importance in reactor design and operation, and because the underlying mechanisms are not fully understood.

The work has been centered about design and construction of a highly instrumented pilot reactor plant with devices for measuring axial and radial temperature profiles and axial concentration profiles. Furthermore, reactor models were developed to simulate the reactor behaviour and experimental runs were performed to obtain data for model discrimination and parameter estimation. The Fischer–Tropsch reaction was chosen as the model reaction in this study.

Testing of the pilot reactor operation showed that the plant performed up to expectations, giving realistic and reliable data. The quality of the analytical results was good as judged from the mass balances.

The range of useful operating conditions was limited due to stability problems typical for this kind of reacting system. The experiments were performed at temperatures about 500 K and partial pressures of synthesis gas between 0.18 and 0.3 MPa, giving conversions in the range 25–50 %. The long term catalyst activity decline observed in the experiments is most likely due to increased diffusional resistance caused by accumulation of liquid products in the catalyst pores.

The kinetic expressions and the estimated optimal parameters gave a reasonable good fit between simulated and experimental concentration profiles. Due to the restricted range of operating temperatures activation energies could not be estimated and typical values from the literature had to be employed. It turned out, however, that the values of the activation energies was not important neither on goodness of fit nor in influence on the values of estimated heat transfer parameters. This was the case for activation energies of CO consumption in the range

80 – 110 kJ/mol and methane formation in the range 110 – 140 kJ/mol, and was due to the small differences in temperature across the bed at the experimental conditions used in this study.

In the traditional dispersion model it is assumed that the void fraction, velocity and thermal conductivity are independent of radial reactor position and that a distinct heat transfer resistance at the wall exists. This model was compared with a physically more realistic model where these parameters were dependent on radial position. Different assumptions concerning the variations of the conductivity in the near–wall region was evaluated for the last model. For all the models heat transfer parameters were estimated from experimental data.

Using the traditional dispersion model, the fluid–mechanical Peclet number was estimated to be about 6.9 which is about 70 % lower than values predicted by available correlations. It is a common observation that effective radial conductivities estimated from measurements under reactive conditions are considerably higher than those obtained from correlations based on cold–flow measurements. This overestimation of the effective conductivity leads to calculated temperature differences which are smaller than the measured ones.

The results from the model with radial dependent parameter values indicated that this model was able to give a more realistic representation of observed temperature profiles in a fixed bed reactor compared to the traditional dispersion model. But the model was rather sensitive to assumptions made on the variations in conductivity in the near–wall region. Since the conductivity is dependent on local velocity, the radial velocity profile is an important factor in determining radial temperature profiles.

Whether a further correction in the conductivity should be taken into account by including a wall function accounting for reflection of stream lines at the wall, remains at the time open. Since the details of the velocity profile in the near–wall region was not well known for the reactor and catalyst particles used in this study, no further conclusions could be made on this question. But the case where the conductivity was assumed proportional to local velocity only gave the best fit between calculated and measured temperature profiles.

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LIST OF SYMBOLS AND ABBREVATIONS

A_{Ki}	Preexponential factor for species i in adsorption constant expressions, 1/MPa
A_i	Preexponential factor for species i in Arrhenius expressions
A'_i	Modified preexponential factor for species i in Arrhenius expressions, $[A_i \exp(-E_i/RT_0)]$
a_v	external particle surface area per unit reactor volume $[6(1-\varepsilon_0)/d_p]$, 1/m
Bi_w	apparent wall Biot number based on particle diameter $[\alpha_w\ d_p\ /\ \lambda_{er}]$, dimensionless
Bi'_w	apparent wall Biot number based on reactor dimensions $[\alpha_w\ R\ /\ \lambda_{er}]$, dimensionless
C	form factor dependent on particle geometry
C, C_1, C_2 ..	general equation coefficients
C_i	concentration of species i in the fluid phase, mol/m^3
C^0_i	initial concentration of species i in the fluid phase, mol/m^3
C_{0i}	inlet concentration of species i, mol/m^3
C_{si}	concentration of species i in the solid phase, mol/m^3
C^0_{si}	initial concentration of species i in the solid phase, mol/m^3

$C^*_{s,i}$	concentration of species i at the surface of a particle, mol/m^3
C_{pg}	heat capasity of the fluid phase, J/(kg K)
C_{ps}	heat capasity of the solid phase, J/(kg K)
CSTR	continously stirred tank reactor
D^0_e	effective diffusivity in stagnant fluid, m^2/s
D_k	Knudsen diffusivity, m^2/s
D_m	molecular diffusivity, m^2/s
D_p	particle effective diffusivity, m^2/s
D_{er}	effective radial diffusivity, m^2/s
D_{ez}	effective axial diffusivity, m^2/s
d_p	catalyst particle equivalent spherical diameter [6·(V_p/S_x)], m
d_t	tube diameter, m
E_i	activation energy for species i, J/mol
FID	flame ionization detector
f_1, f_2	coefficients in equation 2-29
GHSV	gas hourly space velocity, 1/h

ΔH_R	reaction enthalpy, J
ΔHa_i	adsorption enthalpy for species i, J/mol
h_c	heat transfer coefficient between solid and fluid phase, W/(m^2 K)
I_n	Fischer–Tropsch intermediate with n carbon atoms
K_i	adsorption constant for species i, 1/MPa
k_c	mass transfer coefficient between solid and fluid phase, m/s
k_i	rate constant for species i
k_p	rate constant for Fischer Tropsch polymerization reaction
k_t	rate constant for Fischer Tropsch termination reaction
L	reactor length, m
Le	Lewis number $[(C_{ps}\ \rho_p)/(C_{pg}\ \rho_g)]$, dimensionless
l_w	constant in the wall function equation 5–12, units of particle diameters
M	Fischer–Tropsch monomer
Nu_c	fluid–solid Nusselt number $[h_c\ d_p / \lambda_g]$, dimensionless
Nu_w	apparent wall Nusselt number $[\alpha_w\ d_p / \lambda_g]$, dimensionless
P_n	Fischer–Tropsch product with n carbon atoms

P	pressure, MPa
$P_{0,i}$	total pressure at inlet, MPa
P_i	partial pressure of species i, MPa
Pe_h	effective Peclet number for heat based on particle diameter, axial or radial, dimensionless
Pe^0_h	molecular Peclet number for heat based on particle diameter $[d_p \rho_g C_{pg} v_0 / \lambda_g]$, dimensionless
Pe_{hr}	effective radial Peclet number for heat based on particle diameter $[d_p \rho_g C_{pg} v_0 / \lambda_{er}]$, dimensionless
Pe_{hz}	effective axial Peclet number for heat based on particle diameter $[d_p \rho_g C_{pg} v_0 / \lambda_{ez}]$, dimensionless
Pe'_{hr}	effective radial Peclet number for heat based on reactor dimensions, $[R^2 \rho_g C_{pg} v_0 / \lambda_{er} L]$ or $[R^2 \rho_g C_{pg} v / \lambda_r L]$, dimensionless
Pe'_{hz}	effective axial Peclet number for heat based on reactor dimensions, $[L \rho_g C_{pg} v_0 / \lambda_{ez}]$ or $[L \rho_g C_{pg} v / \lambda_{ez}]$, dimensionless
Pe^d_h	fluid–mechanical Peclet number for heat based on particle diameter, axial or radial, dimensionless
Pe^d_{hr}	experimental radial fluid–mechanical Peclet number for heat based on particle diameter, dimensionless.
Pe_m	effective Peclet number for mass based on particle diameter, axial or radial, dimensionless

Pe^{0}_{m} molecular Peclet number for mass based on particle diameter $[d_p\ v_0 / D_m]$, dimensionless

Pe_{mr} effective radial Peclet number for mass based on particle diameter $[d_p\ v_0 / D_{er}]$, dimensionless

Pe_{mz} effective axial Peclet number for mass based on particle diameter $[d_p\ v_0 / D_{ez}]$, dimensionless

Pe'_{mr} effective radial Peclet number for mass based on reactor dimensions $[R^2\ v_0 / D_{er}\ L]$ or $[R^2\ v / D_{er}\ L]$, dimensionless

Pe'_{mz} effective axial Peclet number for mass based on reactor dimensions $[L\ v_0 / D_{ez}]$ or $[L\ v / D_{ez}]$, dimensionless

Pe^{d}_{m} fluid–mechanical Peclet number for mass based on particle diameter, axial or radial, dimensionless

Pe^{d}_{r} theoretical radial fluid–mechanical Peclet number based on particle diameter, dimensionless

Pr Prandtl number $[C_{pg}\ \mu_g / \lambda_g]$, dimensionless

R gas constant, J/mol K

R tube internal radius, m

R_O tube external radius, m

R' ratio of tube internal radius to particle diameter $[R / d_p]$, dimensionless

Re_p	Reynolds number related to the particle diameter $[\rho_g v_0 d_p / \mu_g]$, dimensionless
r	radial coordinate, m
r'	dimensionless radial coordinate [r/R]
r_n	rate of formation of Fischer–Tropsch product with n carbon atoms
$r_{v,i}$	reaction rate of species i, mol/($kg_{catalyst}$ s)
$(-\Delta H)r_v$	total rate of reaction heat generation, W/$kg_{catalyst}$
S_g	catalyst internal surface area, m^2/kg
S_n	Selectivity of Fischer–Tropsch product with n carbon atoms
S_x	external surface area of a particle, m^2
Sc	Schmidt number $[\mu_g / \rho_g D_m]$, dimensionless
T	fluid temperature, K
T^0	initial fluid temperature, K
T_0	fluid temperature at inlet, K
T_s	solid temperature, K
T^0_s	initial solid temperature, K
T^*_s	temperature at the surface of a particle, K

T_c	temperature of the coolant, K
T_w	temperature of the reactor wall, K
TCD	thermal conductivity detector
t	time, s
U_c	overall heat transfer coefficient to the coolant, $W/(m^2 K)$
u	general dimensionless reactor coordinate
V_g	catalyst pore volume, m^3/kg
V_p	volume of a particle, m^3
V_R	reactor volume, m^3
v	local superficial velocity, m/s
v_0	mean superficial velocity, m/s
W	packed bed catalyst mass, kg
X_n	molar fraction of hydrocarbons with n carbon atoms relative to C_1
x	catalyst particle coordinate, m
x'	dimensionless catalyst particle coordinate $[2x/d_p]$
y	dependent variable in reactor model

y_i	dimensionless concentration of species i in the fluid phase, $[C_i / C_{0,i}]$
z	axial coordinate, m
z'	dimensionless axial coordinate $[z/L]$

Greek letters

α	probability of chain growth in Fischer–Tropsch synthesis
α_c	heat transfer coefficient at the coolant side, W/(m^2 K)
α_w	heat transfer coefficient at the wall, W/(m^2 K)
β_i	linear rate of deactivation for species i, 1/h
γ	molar C_2/C_3 ratio
ε	local void fraction of the packed bed, dimensionless
ε_w	void fraction at the interior of the packed bed, dimensionless
ε_0	mean void fraction of the packed bed $[1-(\rho_b/\rho_p)]$, dimensionless
ε_p	void fraction of catalyst particle, dimensionless
η	catalyst effectiveness factor
λ^0_e	effective thermal conductivity of bed with stagnant fluid, W/(m K)
λ^d_e	dynamic contribution to effective thermal conductivity, axial or radial W/(m K)

λ_r	radial effective thermal conductivity of bed depending on position, W/(m K)
λ_{rr}	mean radial effective thermal conductivity of bed, W/(m K)
λ_{rz}	axial effective thermal conductivity of bed, W/(m K)
λ_g	thermal conductivity of the fluid phase, W/(m K)
λ_p	effective thermal conductivity of a catalyst particle, W/(m K)
λ_w	thermal conductivity of the reactor wall, W/(m K)
μ_g	viscosity of the fluid phase, N s/m^2
θ	dimensionless fluid temperature, [T/T$_0$]
θ_c	dimensionless coolant temperature, [T$_c$/T$_0$]
ρ_b	catalyst bulk density, kg/m^3
ρ_g	density of the fluid phase, kg/m^3
ρ_p	catalyst particle density, kg/m^3
ρ_s	catalyst solid density, kg/m^3