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Modeling and simulation of a small-scale trickle bed reactor for sugar hydrogenation

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ABSTRACT

Laboratory-scale trickle bed reactor was modeled and simulated, taking into account axial dispersion, gas–liquid, liquid–solid and internal mass transfer as well as catalyst deactivation under isothermal conditions. For catalyst particles dynamic and steady state models were developed, including both mass and heat balances. Catalyst deactivation was included in the model by using the final activity concept for the catalyst particles. A well-working numerical algorithm (method of lines) was applied for solving the reactor model with Matlab 7.1 and the results followed experimental trends very well. The steady-state reactor model was based on simultaneous solution of mass balances. The aim was to illustrate how these parabolic partial differential equations could be solved with a step-by-step calculation for a selected geometry. The final model verification was done against experimental data from the hydrogenation of arabinose to arabitol on a ruthenium catalyst.

1. Introduction

Trickle beds are among the most popularly used three-phase reactors in industrial scale. These kinds of reactors are used in oil refining as well as in chemical and petrochemical industry. A trickle bed reactor is a packed bed filled with a layer of porous catalyst particles through which gas and liquid flows typically co-currently downwards. Strong interaction between the hydrodynamics (flow conditions), mass transfer effects and intrinsic reaction kinetics make the mathematical modeling of these kinds of reactors challenging. Various flow regimes may prevail in the catalytic packed bed, depending on the superficial gas and liquid velocities and operating conditions (pressure, temperature, detailed packed bed geometry etc.). Small scale trickle-bed reactors are useful for generating information from existing processes, and especially, for studying new technically and economically attractive catalysts. Various effects such as incomplete wetting, deviations from plug flow, channeling inside the catalyst bed and backmixing of liquid (which all otherwise add challenges to the modeling) can all to a limited extent be reduced by proper dilution of the catalyst bed with fine inert particles.

Modeling of trickle bed reactors (TBR) has been reviewed in several textbooks (Gianetto & Silveston, 1986; Ramachandran & Chaudhari, 1983; Ranade, Chaudhari, & Gunjal, 2011; Trambouze et al., 1988). Here, different models are discussed and simulation results for hydrogenation of L-arabinose to arabitol are given. The raw material, L-arabinose, can be obtained from biomass, for instance, from the hemisellulose arabinogalactan, which appears in wood, particularly in *Larix sibirica*. Reaction kinetics, mass transfer, flow related effects and deactivation were included. Commercial Ru/C served as the catalyst. The case is an example of an environmentally friendly route to produce an alternative sweetener and food additive. The aim of this study was to give insight in the effects of various phenomena present in a laboratory-scale TBR. The isothermal reactor modeling part is totally based on the work and findings of Durante (2010).

2. Methods

2.1. Sugar hydrogenation kinetics in slurry reactor

2.1.1. Experimental arrangement

Semi-batch hydrogenations of L-arabinose were conducted (Durante, 2010) in a laboratory-scale slurry reactor (T = 90–130 °C,

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Symbols

a	specific surface area [m^2/m^3]
A_{Arr}	Arrhenius constant in reaction rate expression, convenient units to produce reaction rate as [mol/(ls)]
A, B	parameters of the step-by-step temperature calculation [W/m^3K]
A_p	surface area of the particle [m^2]
A_R	cross-sectional surface area of the reactor [m^2]
C_1	parameter of the step-by-step temperature calculation [W/m^3K]
C_i	local concentration of component i in liquid, $i = H_2$, arabinose, arabitol, [mol/l]
$C_{H_2,L*}$	solubility of component H_2 in liquid [mol/l]
$C_{i,L}$	concentration of component i in liquid, $i = H_2$, arabinose, arabitol[mol/l]
$C_{i,IN}$	concentration of component i in liquid feed, $i = H_2$, arabinose, arabitol, [mol/l]
$C_i^m C_i^{m-1}$	concentration in location i at time steps m and $m-1$
$C_{i,s}$	concentration of component i at surface, $i = H_2$, arabinose, arabitol [mol/l]
C_s	surface concentration [mol/l]
D_1	parameter in step-by-step calculation routine [K]
D_a	axial dispersion coefficient in liquid [m^2/s]
D_{eff} $D_{eff,i}$	effective diffusivity, of the compound i, $i = H_2$, arabinose, arabitol [m^2/s]
D_i	liquid and effective diffusivity of component $i = A$, B, H_2 [m^2/s]
$D_{i,L}$	diffusivity of component i in liquid [m^2/s]
$d_{p,inert}$	inert particle diameter [m]
d_p	particle diameter [m]
d_R	diameter of the reactor [m]
E	parameter in step-by-step calculation [W/m^3K]
E_A	activation energy, of the main reaction [J/mol]
f_s	relative saturation [ ]
$f_{A,x-1}$, $f_{A,x}$, $f_{A,x+1}$	dimensionless concentration for arabinose before, at and after step x [ ]
$f_{A,1}$, $f_{A,2}$, $f_{A,N}$	dimensionless concentration for arabinose in first second and last step [ ]
$f_{H_2,x-1} f_{H_2,x} f_{H_2,x+1}$	dimensionless concentration for hydrogen before, at and after step x [ ]
$f_{i,x-1} f_{i,x} f_{i,x+1}$	dimensionless concentration of component i ($i = H_2$, A, B), in step $x-1$, x and $x+1$
H_{H_2}	Henry's law constant for H_2 [Pa]
k	reaction rate constant, units to produce rate as [mol/(ls)]
k_1, k_2	modified reaction rate constant for main and coking reaction, units to produce rate as [mol/(ls)]
k'	deactivation rate constant units to produce activation change as [1/s]
k_i	reaction rate constant for reaction i, units to produce rate as [mol/(ls)]
$k_{GL}a$	united liquid–solid mass transfer coefficient, for hydrogen [1/s]
$k_{A,LS}a$	united liquid–solid mass transfer coefficient, for arabinose [1/s]
$k_{B,LS}a$	united liquid–solid mass transfer coefficient, for arabinose [1/s]
$k_{H_2,LS}a$	united liquid–solid mass transfer coefficient, for hydrogen [1/s]
K_A, K_{H_2}, K_B	adsorption parameters for glucose, hydrogen and sorbitol [l/mol]
K_i	adsorption parameter for component $i = A$, B, H_2 [l/mol]
L	location [m]
L_R	reactor length [m]
m_{cat}	mass of catalyst [kg]
m	time moment [ ]
n	reaction order [ ]
N	total number of volume elements [ ]
p	pressure [Pa]
p_{H_2}	partial pressure of H_2 [Pa]
Pe	Peclet number [ ]
Q_L	volumetric flow rate of liquid [ml/min]
r, r_p, r	radial location, in particle, in catalyst bed [m]
$r_{0,S}$	power law part of rate for component I at surface [mol/(ls)]
R, R_i	reaction rate, for component i, $i = H_2$, A (arabinose), B (arabitol) [mol/(ls)]
R_1, R_2	reaction rate of the main and coking reaction [mol/(ls)]
R_S	reaction rate of main reaction at the surface of the particle [mol/(ls)]
r_p	particle radius [m]
r_p	characteristic radius of particle [m]
R_g	general gas constant [J/(mol K)]
s	surface shape factor [ ]
T	temperature [°C]
T, T_{x-1}, T_x, T_{x+1},T_S	temperature at step $x-1$, x, and $x+1$ and at surface [°C] or [K]
t	time, [s]
v_i	stoichiometric coefficient for reaction [ ]
V_p, V_x	volume of the particle and volume element x [m^3]
w_G	superficial gas velocity [m/s]
w_L	superficial liquid velocity [m/s]
x	dimensionless location [ ]
Y	parameter for step-by-step computation of concentration profile
z	location [m]
X	parameter for step-by-step computation of concentration profile
X_A	conversion of arabinose [ ]
Z_i	parameter for component $i = A$, B, H_2 [1/(mol/ls]
Δt	time step [s]
Δx	dimensionless step length [ ]
z	location [m] or dimensionless location [ ]
α, α_0, α^*	activity, final activity [ ]
β	conversion factor
ΔH_r	reaction enthalpy [J/mol]
ε	volume fraction [ ]
ε_p	particle porosity [ ]
ε_L	liquid hold-up [ ]
ε_B	porosity of the catalyst bed [ ]
η	effectiveness factor for component [ ]
θ_c	occupation factor for coke [ ]
λ	thermal conductivity [W/mK]
μ_l	viscosity of liquid [kg/(ms)]
$\rho_L \rho_G$, ρ_p	density of liquid, gas and particles [kg/m^3]
ρ_B	catalyst mass concentration [kg_{cat}/m^3]
τ_p	tortuosity [ ]
Φ	Thiele modulus [ ]