MODELING OF DYNAMIC BEHAVIOUR OF TRICKLE-BED REACTOR

David Skála[a],*Petra Houserová*[b],*Jiří Hanika*[c]

Department of Organic Technology, ICT Prague, Technická 5, 16628 Prague 6

a….David.Skala@vscht.cz
b….Petra.Houserova@vscht.cz
c….Jiri.Hanika@vscht.cz

Introduction:

Trickle bed reactors with gas-liquid co-current downflow have been widely used in hydrogenation and hydrodesulfurization processes. Recent published papers [1-4] dealing with periodically operated trickle bed reactors for catalytic reactions propose that the periodic flow interruption can generate a higher conversion than the steady state operation.

The aim of this study was focused on modeling of flow interruption feed in trickle bed. Two models of trickle bed reactor were solved a) steady state model and b) pseudo-steady state. This work is important for providing a theoretical basis for experimental work aimed to find optimum conditions, for trickle-bed reactor operation under dynamic conditions. Numerical solution of the model and results visualization were provided using the software Matlab, version 5.3.

Character of periodic operation:

The variation of a liquid film thickness covering external surface of catalyst pellets is a typical feature of the periodic trickle bed operation under such conditions the lower mean value of the value of flowing liquid film thickness can improve the gas component transfer to the catalyst active sites. Starting the model formulation there is necessary to define some parameters, characterizing the system. These parameters follow from the situation in Figure 1 and are represented by the following symbols.

LP-period length, time between beginning of one FEED ON cycle to the next FEED ON

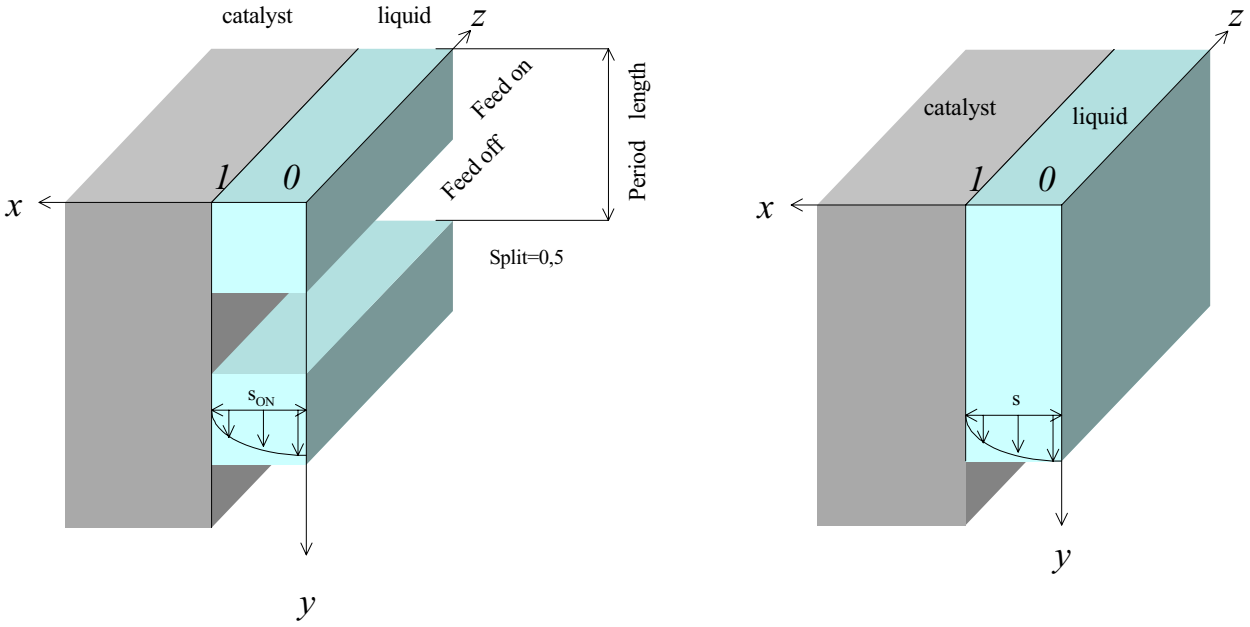
S -split, length of FEED ON in one period

Assumption of the pseudosteady state model :

Trickle bed is replaced by a single large catalyst pellet. Liquid reaction mixture and the surface of the liquid film flowing along the external surface of the catalyst, are assumed to be saturated by the gaseous component to the gas-liquid equilibrium concentration value. Pseudosteady state model supposing variable liquid film thickness along the catalytic wall is schematically shown in left part of Fig.1. The right part of this figure illustrates the steady state model with constant liquid film thickness. Both models were formulated using the following assumption.

The concentration profile of the gaseous component dissolved in the film is given by the pertinent parabolic partial differential equation supposing convective and diffusion transport in longitudinal and transversal directions, respectively . The equation is based on the mass and momentum balance of a space element within the film.

Figure 1. Pseudo-steady state and steady regimes of the model trickle bed reactor



The steady state and pseudo-steady state models, for isothermal regime of reactor, were formulated and solved in this study. The liquid flow was assumed to be a laminar one. Numerical simulations were made for a simple, irreversible, monomolecular, first order

reaction with respect to a gas component dissolved in the liquid. The reaction took place on the external surface of catalyst wall.

Model formulation:

The following partial differential equation was used for description of the dynamics of the diffusion transport across the film and convective mass transport in the flow direction. Model supposes the parabolic velocity profile in the flowing liquid film.

$$\partial c/\partial T=\partial^2 C/\partial X^2-Pe*P*(1-X^2)*\partial c/\partial Y \quad (1)$$

The equation can be simplified for the steady state of the system

$$0=\partial^2 C/\partial X^2-Pe*P*(1-X^2)*\partial c/\partial Y \quad (2)$$

Dimensionless time T is defined by

$$T=t*D/s_{SS}, \quad (3)$$

Where D is diffusion coefficient of dissolved gas in the liquid.

Peclet number is represented by the following definition

$$Pe=w_{max*}s_{SS}/D \quad (4)$$

w_{max} is external surface velocity in the flowing liquid film

$$w_{max}=\rho* g* s_{SS}^2/2\mu \quad (5)$$

Geometric simplex of reactor is defined by the relation.

$$P=s_{SS}/L, \quad (6)$$

Here C stands for dimensionless concentration, defined as ratio

$$C = c/c^0. \quad (7)$$

Dimensionless coordinates X and Y were defined by the following relations

$$X=x/s_{SS} \quad (8)$$

$$Y=y/L \quad (9)$$

Symbols s_{ss} and L represents the film thickness and length, respectivelly.

Because the reaction mixture is assumed to be saturated with gas, e.g. hydrogen in case of hydrogenation, the inlet concentration was equal to unity. The following initial and boundary conditions were applied for the model solution:

Initial conditions: T=0:

$$C(X,0)=1 \quad (10)$$

$$C(X,Y)=0. \quad (11)$$

Boundary conditions:

$$Y=0 \quad C=c^0 \quad (12)$$

$$Y=1 \quad \partial c/\partial Y = 0 \quad (13)$$

$$X=0 \quad C=c^0 \quad \text{l-g interface} \quad (14)$$

$$X=1 \quad \partial c/\partial X= -Th*c \quad \text{s-l interface} \quad (15)$$

where Th is modified dimensionless Thiele modulus

$$Th=K.s_{SS}/D. \quad (16)$$

The steady state liquid film thickness s_{ss} can be calculated from the formula

$$s=(3 G \mu / \rho^2 g)^{1/3} \quad (17)$$

Where G stands for specific liquid wetting mass velocity , μ and ρ represents viscosity and density of liquid.

Model solution

Periodic feed variations cause changes of both parameters Pe*P and Theta. It is necessary to characterize these changes. It is obvious, that Pe*P product is depended to film thickness powered to 4 and on the other hand Thiele modulus is proportional to film thickness. Film thickness depends on mass velocity G.

During feed-on part of the model parameters can be calculated from parameters values representing the feed-off part of period in which the feed was supposed to be ten times smaller comparing to the feed-on regime.

$$PePon=(10)^{\wedge}4/3*PePoff \quad (18)$$

$$Thetaon=(10)^{\wedge}1/3*Thetaoff \quad (19)$$

For constant average feed holds the following relations:

$$PePss=((10+1)/2)^{4/3}*split*PePoff \quad (20)$$

$$Thetass=((10+1)/2)^{1/3}*Thetaoff \quad (21)$$

The variables of interests include Thiele modulus, Peclet number and geometric simplex. Table 1 brings interval of variables values of interest.

Table 1. Reaction system parameter values used for model solution in comparison to the experimental ones

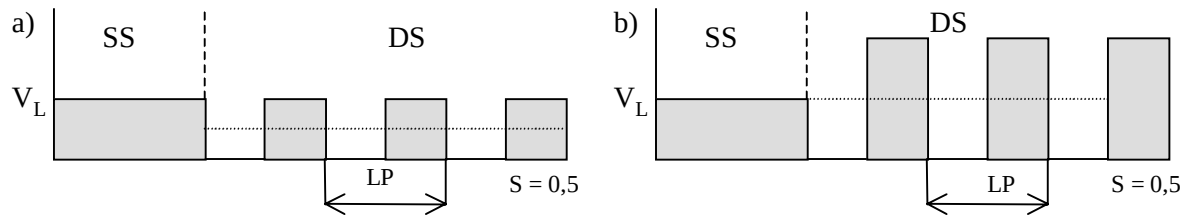
Parameter	**Model**	**Experimental Value**
Theta	0.1-100	1.02
Pe*P	0.1-100	0.07
K [cm/s]	-	$1.7*10^{-2}$-$3*10^{-2}$
s	-	0.022 mm

Experimental

Simulation results were compared with the results of laboratory experiments. Cyclohexene hydrogenation on the Cherox 41-00 (3% Pd on charcoal) was chosen as a model reaction. Two types of experiments were provided **a)** with constant bed irrigation by liquid feed in dynamic and steady state regimes, **b)** with constant time average feed in dynamic and steady state regimes. Figure 2 shows schematically these two variants.

Figure 2. Feed rate changes in steady (SS) and dynamic state (DS) regimes

a) Constant bed irrigation b) constant time mean feed rate



Results and discussion

The average outlet concentration values for constant feed and/or, forced feed oscillations were compared in this study. The ratios of average concentration in forced periodic regime and steady state concentration is given in figures 3 and 4.

Figure 3. Comparison of steady-state and pseudo-steady state efficiency.
Constant time mean feed rate

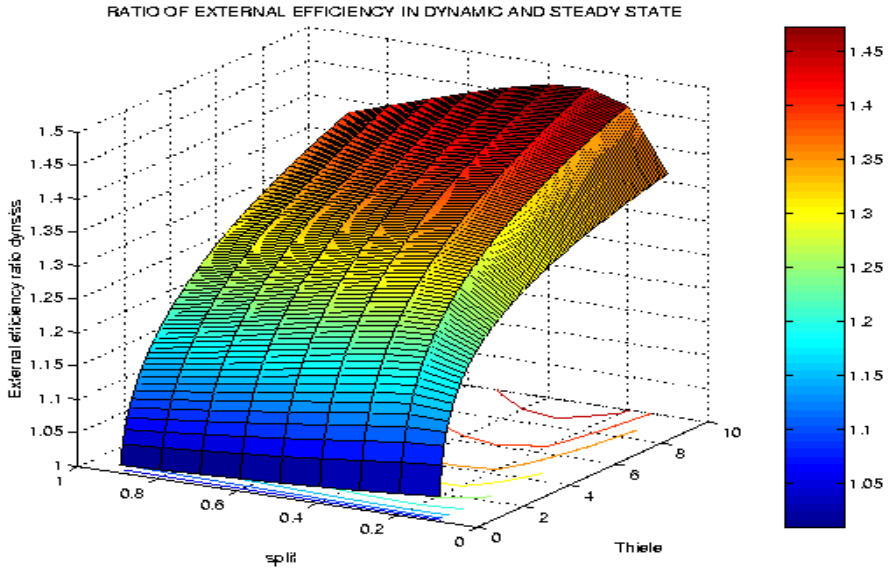
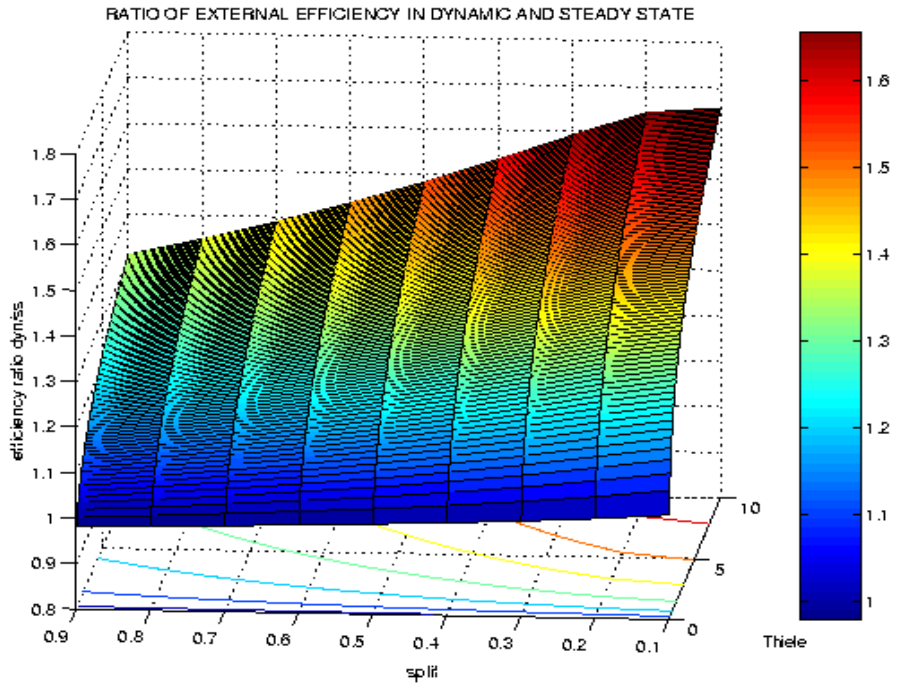


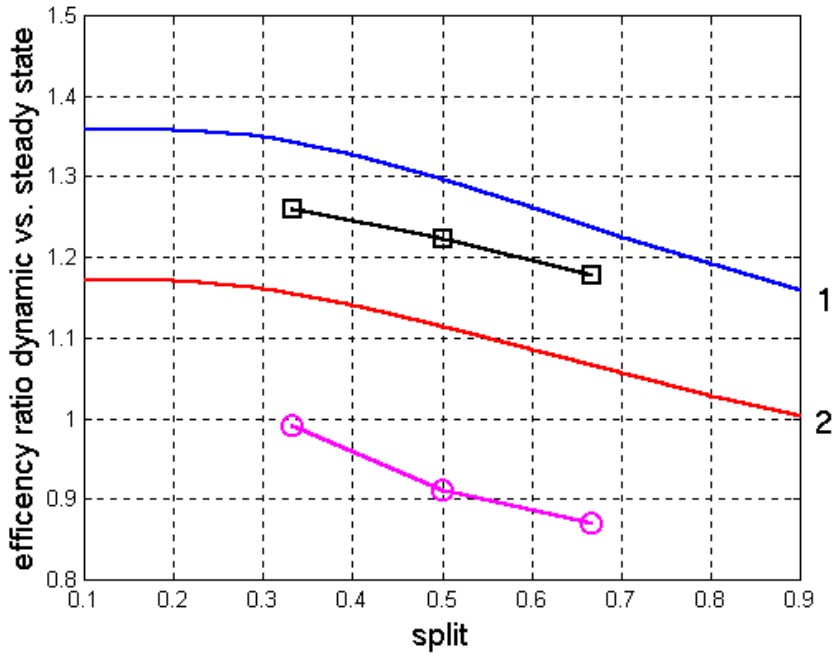
Figure 4. Comparison of steady-state and pseudo-steady state efficiency.
Constant bed irrigation



Experimental results verified the model solution resulting in the fact, that the periodic feed interruption can lead to reactor performance enhancement. It was noticed that the biggest performance enhancement was reached at the very short split value.

The comparison of the model solution and experimental data is given in the figure 5.

Figure 5. Influence of cycle split on reactor performance



Curve 1 represents numerical results for constant bed irrigation.

Curve 2 represents numerical solution for constant time mean feed rate.

Square points correspondent to experimental data for constant bed irrigation and circle ***points*** represent experimental results measured at constant time mean feed rate.

Conclusion

Experimental results verified strong effect of split value on a mean outlet stream conversion. The best reactor performance was reached at the very short split value. This fact is in agreement with numerical results, which manifest the same trend. It's possible to say that qualitative agreement between numerical and experimental data was reached in this study. But further experiments and more sophisticated models describing the dynamic behavior of the trickle bed reactor are necessary for better understanding the very complex phenomena in the system.

Nomenclature:

c - concentration (-)
C - dimensionless concentration (-)
D - diffusion coefficient (m^2/s)
G - specific mass velocity ($g\ m^{-1}\ s^{-1}$)
K - reaction rate coefficient ($m^3 m^{-2}_{kat} s^{-1}$)
L - effective reactor length (m)
LP - period length (s)
P - geometric simplex (-)
Pe - Peclet number (-)
s - liquid film thickness (m)
S - split (-)
t - time (s)
T - dimensionless time (-)
w_{max} - external surface liquid velocity ($m\ s^{-1}$)
x - coordinate (m)
X - dimensionless coordinate (-)
y - coordinate (m)
Y - dimensionless coordinate (-)
Theta - Thiele modulus (-)
μ - viscosity ($g\ s^{-2}$)
ρ - density ($kg*m^{-3}$)

Indexes:
upper:
0 - initial value
lower:
ON - dynamic state, set on
OFF - dynamic state, set off
SS - steady state

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