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Modelling of reactive separations including fast chemical reactions in CSTR

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Abstract

Reaction and separation in situ brings synergic effect of various phenomena (kinetic, heat and mass transfer) and phase equilibria. When modelling such a process, various types of models can be used.

The goal of this paper was to create a non-equilibrium mathematical model of a reactor, in which reactive distillation or absorption with fast chemical reaction takes place. To characterize the vapour–liquid contact, film layer theory was used. In the presented paper, Fick's equation is used to describe mass transfer with reaction in the liquid film. It is suitable for systems where a fast reaction occurs in the liquid film.

Simulation of a steady-state reactive distillation in a CSTR with a partial condenser is presented. The simulation code was developed in FORTRAN. Steady-state modelling leads to a system of both non-linear algebraic and second-order non-linear differential equations. Solution of the system is provided by the imported solver BUNLSI. Within every iteration a boundary value problem (reaction and mass transfer in the liquid phase) had to be solved with a solver from the IMSL mathematical library. The influence of the Hatta number on the CSTR performance was investigated for a simple second-order reversible reaction.

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Keywords: Reactive distillation; Reactive absorption; Film model; Fast reaction; Mathematical model

1. Introduction

Implementation of reactive separations brings many benefits. According to Taylor and Krishna [1], advantages of reactive distillation (RD) are as follow: vast capital savings caused by merger of the reactive and separation equipment, conversion enhancement up to 100%, better selectivity, lower catalyst requirements, possibility of azeotrope "overlap", reaction heat utilization in components separation, etc. Disadvantages of such processes are, e.g. volatility constrains, residence time requirements or scale up to larger flows. The best performance appears when using RD for esterification, etherification, alkylation and hydrolysis.

Two main approaches can be used in mathematical model development. Equilibrium models assume thermodynamic equilibrium between liquid and vapour leaving the reactive distillation CSTR or column's tray. Non-equilibrium models involving the film theory assume that phase equilibrium takes place only on the vapour–liquid interface. A thin film exists on both sides of the interface representing significant resistance against the heat and mass transfer. When dealing with reactive absorption or very fast homogenous reactions, chemical reactions in the liquid film have to be taken into account. Mass transfer in the film can be described by various equations. Taylor and Krishna [1] suggest the Maxwell–Stefan (MS) [2] approach for rate-based modelling of the RD processes. In the work of Frank et al. [3] the authors use the MS equations for modelling simultaneous reaction (a reversible one) and mass transfer in the liquid film with fixed film's boundary conditions and compare the results to those obtained using the Fick's law. As a conclusion they strongly advise to use the MS approach also for homogeneously catalysed reactions (simultaneous mass transfer and chemical reaction) even despite the fact that for very fast chemical reactions numerical problems arise. The comparison of both approaches is summarized in Table 1. Many authors (from numerous available papers in literature cf. [4,5], and also in our papers [6,7]) described the V–L interface using the MS equations especially

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Nomenclature

a_v	specific gas–liquid interfacial area ($m^2\,m^{-3}$)
c	molar concentration ($mol\,m^{-3}$)
D_ℓ	diffusion coefficient in the liquid phase ($m^2\,s^{-1}$)
$\dot{D}$	distillate molar flow ($mol\,s^{-1}$)
$\dot{F}$	feed molar flow ($mol\,s^{-1}$)
h	molar enthalpy ($J\,mol^{-1}$)
Ha	Hatta number, defined in Eq. (19)
k_G	gas-side mass transfer coefficient ($mol\,m^{-2}\,s^{-1}\,Pa^{-1}$)
k_ℓ	liquid-side mass transfer coefficient ($m\,s^{-1}$)
k_V	reaction rate constant, dimension depends on the reaction type
$k_{V\infty}$	frequency factor, dimension depends on the reaction type
K_C	equilibrium constant of chemical reaction
$\dot{L}$	molar flow of the liquid reflux ($mol\,s^{-1}$)
$\dot{N}$	molar flux ($mol\,m^{-2}\,s^{-1}$)
N_I	number of components
N_R	number of chemical reactions
P	pressure (Pa)
$\dot{Q}$	heat input/output flow ($J\,s^{-1}$)
T	temperature (K)
$\dot{V}$	molar flow of vapours leaving the CSTR ($mol\,s^{-1}$)
V_R	volume of the bulk liquid in the reactor (m^3)
$\dot{W}$	molar flow of the reactor liquid output ($mol\,s^{-1}$)
x	molar fraction in the liquid phase
y	molar fraction in the gaseous phase
z	spatial coordinate in the liquid film (m)

Greek letters

$\Delta_r H$	reaction enthalpy ($J\,mol^{-1}$)
δ	liquid film width (m)
γ	activity coefficient in the liquid phase
ν	stoichiometric coefficient
$\dot{\xi}_V$	rate of chemical reaction ($mol\,m^{-3}\,s^{-1}$)

Subscripts

c	condenser
del	delivered
D	distillate
f	liquid film
F	feed
i	component index
j	reaction index
k	key component
ℓ	liquid phase
L	liquid reflux from condenser
r	reaction
V	vapour outlet of the CSTR
W	liquid outlet of the CSTR
z	position in the film

Superscripts

ℓ	liquid phase
g	gaseous phase
$\circ$	pure component
$*$	interface equilibrium

Table 1
Pros (+) and cons (−) of the Maxwell–Stefan and Fick approaches for simulation of liquid film with homogeneous reactions

Maxwell–Stefan	Fick
+ More exact results	+ Simple
− Much more complicated	+ Reaction source can be easily added
− Many parameters are needed	+ No additional parameters are needed
− Reaction source cannot be easily added	+ Possible non-uniform discretization
− Usually leads to uniform discretization	− Less exact when concentration non-idealities occur
− Low numerical stability if a very fast chemical reaction occurs	

when modelling heterogeneously catalysed reactions, e.g. no reactions in the liquid film. The Fick's law is predominantly used for a G–L reactor or reactive absorption [8,9,10].

In this work, the non-equilibrium reactive distillation or absorption model of a continuous stirred tank reactor (CSTR) with a partial condenser (schematically shown in Fig. 1) with a very fast homogeneous chemical reaction running in the reactor's bulk liquid and in the liquid film is presented. The reaction–diffusion problem on the vapour–liquid interface has been solved using Fick's law. In the paper algorithm of solution, even for very high chemical reaction rates, is proposed using non-uniform discretization of model equations in the liquid film.

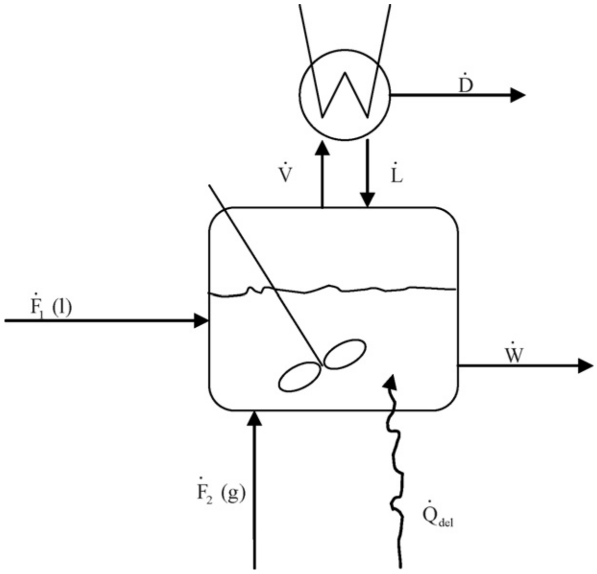

Fig. 1. Scheme of a CSTR with a partial condenser.

Influence of the Hatta number on the reactor performance has been investigated in the simulations.

2. Mathematical model

The basic assumptions taken into consideration are as follows:

- Temperature of both bulk phases and films on the gas–liquid interface is the same.
- Reaction takes place only in the liquid phase (liquid film and bulk liquid).
- Constant pressure in the system.
- Partial condenser.
- Ideal behaviour of the gaseous phase.
- Thermodynamic equilibrium only on the interface alone.
- Steady state.

The mathematical model presented in the following text consists of three parts. The first one is made of equations describing mass transfer and chemical reaction in the film. The second one represents a model of the reactor which consists of material balances of all components in each phase separately and of the overall enthalpy balance of the reactor. The model is completed by equations characterizing the condenser.

To solve the material balances, values of components' molar fluxes through the vapour–liquid interface are required (Fig. 2). These values can be obtained by solving reaction–diffusion equations in the liquid film. To solve the liquid film model equations, both bulk liquid and gas phase concentrations are needed as boundary conditions for the reaction–diffusion equation.

2.1. Film model

To describe the reaction and diffusion in the film, Fick's equation has been used since fast reaction in the film cannot be neglected. Thus, material balance of the *i*th component can be written as follows:

$$D_{\ell,i}\left(\frac{\mathrm{d}^2 c_{f,i}}{\mathrm{d}z^2}\right) = -\sum_{j=1}^{N_\mathrm{R}} \nu_{i,j}\dot{\xi}_{V,j} \tag{1}$$

with boundary conditions (BC's):

$$\begin{aligned} z = 0: &\quad c_{f,i} = c^*_{f,i} \\ z = \delta: &\quad c_{f,i} = c_{\ell,i} \end{aligned} \tag{2}$$

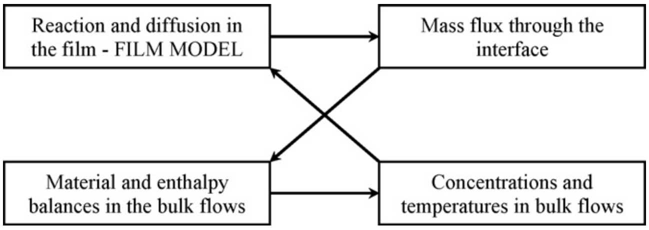

Fig. 2. Basic structure of the mathematical model.

As was mentioned above, to solve bulk material balances of the reactor, interface molar fluxes are needed. The molar flow of the *i*th component entering the gaseous phase can be calculated from the concentration gradient in the liquid film at $z = 0$, i.e.:

$$(\dot{N}_i)_{z=0} = D_{\ell,i}\left(\frac{\mathrm{d}c_{f,i}}{\mathrm{d}z}\right)_{z=0} \tag{3}$$

for which, considering no reaction in the gaseous film, can be written in the following form:

$$(\dot{N}_i)_{z=0} = k_{\mathrm{G},i}(\gamma_i P_i^\circ x^*_{f,i} - P_i) \tag{4}$$

assuming the vapour–liquid equilibrium on the interface being described by equation:

$$P y_i^* = P_i^\circ \gamma_i x^*_{f,i} \tag{5}$$

The molar flux of a component entering the bulk liquid phase is given by the concentration gradient in the liquid film at $z = \delta$:

$$(\dot{N}_i)_{z=\delta} = -D_{\ell,i}\left(\frac{\mathrm{d}c_{f,i}}{\mathrm{d}z}\right)_{z=\delta} \tag{6}$$

The thickness of the liquid film, δ, depends on the reactor hydrodynamic conditions and physical–chemical properties of the liquid phase [2,10]. Conventionally it is defined as the ratio of the diffusion coefficient and the liquid-side mass transfer coefficient for the chosen key component:

$$\delta = \frac{D_{\ell,k}}{k_{\ell,k}} \tag{7}$$

2.2. Reactor model

The model of a continuous stirred tank reactor has been used to describe the reactor, with the gaseous and bulk phases treated separately. Material balance of the *i*th component in the bulk liquid phase is:

$$\dot{F}_i^\ell + V_\mathrm{R}\sum_{j=1}^{N_\mathrm{R}} \nu_{i,j}\dot{\xi}_{V,j} + \dot{L}x_{\mathrm{c},i} - V_\mathrm{R} a_\nu (\dot{N}_i)_{z=\delta} - \dot{W}x_i = 0 \tag{8}$$

Material balance of the *i*th component in the bulk gaseous phase:

$$\dot{F}_i^\mathrm{g} + V_\mathrm{R} a_\nu (\dot{N}_i)_{z=0} - \dot{V} y_i = 0 \tag{9}$$

If chemical reactions in the system do not have a significant thermal effect, temperature can be assumed to be equal in all bulk phases and films. The overall reactor's enthalpy balance can be written (reference point for enthalpy calculations—liquid phase, pure component at reference temperature T^0) in this form:

$$\begin{aligned} &\sum_{f=1}^{N_{F\ell}} \dot{F}_f^\ell h_F^\ell + \sum_{f=1}^{N_{F\mathrm{g}}} \dot{F}_f^\mathrm{g} h_F^\mathrm{g} + \int_0^\delta \sum_{j=1}^{N_\mathrm{R}} \Delta_\mathrm{r} H_j \dot{\xi}_{V,j}\,\mathrm{d}z \\ &\quad -V_\mathrm{R}\sum_{j=1}^{N_\mathrm{R}} \dot{\xi}_{V,j}\Delta_\mathrm{R} H_j + \dot{Q}_\mathrm{del} + \dot{L}h_L - \dot{V}h_D - \dot{W}h_W = 0 \end{aligned} \tag{10}$$

The mathematical model of the reactor is completed by the summation equations:

$$\sum_{i=1}^{N_I} x_i = 1 \tag{11}$$

$$\sum_{i=1}^{N_I} y_i = 1 \tag{12}$$

2.3. Model of the condenser

A partial condenser was assumed. Either the temperature (T_c) or the heat output ($\dot{Q}_c$) can be chosen as a fixed parameter. The condenser has been modelled as an equilibrium one because of a very good contact between the vapour and liquid phase without a chemical reaction. The overall material balance can be written as follows:

$$\dot{V} y_i - \dot{L} x_{c,i} - \dot{D} y_{c,i} = 0 \tag{13}$$

The enthalpy balance then has the following form:

$$\dot{V} h_V - \dot{L} h_L - \dot{D} h_D - \dot{Q}_c = 0 \tag{14}$$

An equilibrium is assumed between the liquid and vapour condenser outlets:

$$P y_{c,i} = P_i^{\circ} x_{c,i} \gamma_i \tag{15}$$

Again, the summation equations (Eqs. (11) and (12)) were used.

2.4. Model system solution

The model equations represent a mixed system of:

- strongly non-linear algebraic equations (NAE) describing the material and enthalpy balances in the bulk phases of the reactor and condenser (Eqs. (8) and (9));
- second-order ordinary differential equations with boundary conditions in two points (BVP problem) describing reaction and diffusion in the liquid film (Eqs. (1)–(6)).

A FORTRAN code has been built to simulate the reactor. To solve NAE, an imported solver, BUNLSI [11] was used. Within every iteration of this solver the ODE's were solved by the solver DBVPFD from IMSL mathematical library [12], which uses automatically adjustable non-uniform discretization for the space variable. This enables avoiding numerical problems connected with very fast chemical reactions between the reactant entering the liquid film from the gaseous phase and the reactant present in the liquid phase.

3. Model system

The assumed model consists of four components taking part in a reversible reaction:

$$\mathrm{A} + \mathrm{B} \leftrightarrows \mathrm{R} + \mathrm{S} \tag{R1}$$

Table 2
Selected parameters of the model

N_I	4
N_R	2
P (Pa)	101,325
V_R (m^3)	3
k_ℓ (m s^{-1})	1.5×10^{-4}
k_G (mol m^{-2} s^{-1} Pa^{-1})	1.5
a_ν (m^2 m^{-3})	10–100
$k_{V\infty 1}$ (m^3 mol^{-1} s^{-1})	3.97×10^{-8}
$k_{V\infty 2}$ (m^3 mol^{-1} s^{-1})	1.35×10^{-8}
E_{A1} (J mol^{-1})	5000
E_{A2} (J mol^{-1})	5000

which can be rewritten as two irreversible reactions:

$$\mathrm{A} + \mathrm{B} \rightarrow \mathrm{R} + \mathrm{S} \tag{R2}$$

$$\mathrm{R} + \mathrm{S} \rightarrow \mathrm{A} + \mathrm{B} \tag{R3}$$

The reaction rate of (R1) can be generally written in the following form:

$$\dot{\xi}_V = k_{V1}\left(c_A c_B - \frac{c_R c_S}{K_c}\right) \tag{16}$$

or if (R1) is rewritten in the form of (R2) and (R3), respective reaction rates are following:

$$\dot{\xi}_{V1} = k_{V1} c_A c_B \tag{17}$$

$$\dot{\xi}_{V2} = k_{V2} c_R c_S \tag{18}$$

One of the reactants (B) was fed in gaseous phase. To calculate activity coefficients in the liquid phase (γ) the Wilson equation has been chosen. Initial values of the kinetic parameters have been used as stated in Table 2. Physical and chemical properties have been modelled for a system of ethyl acetate esterification and were taken out of the HYSYS® commercial software database and [13], with respect to the following naming: A = acetic acid; B = ethanol; R = ethyl acetate; S = water.

4. Results and discussion

In order to illustrate the capability and robustness of the developed simulation code, the simulation of reactive distillation in a CSTR with a partial condenser using the reactive system described in the previous section is presented and discussed here. To investigate the influence of mass transfer resistance and fast chemical reaction rate in the liquid phase on reactor behaviour, the Hatta number is used, defined as:

$$Ha^2 = \frac{\dot{\xi}_{V,j} D_{\ell,k}}{k_{\ell,k}^2 c_{\ell,k}} \tag{19}$$

Subscript k indicates the chosen key component (in our case component B), and j the principal chemical reaction (R1), $c_{\ell,k}$ is the bulk concentration of the key component in the bulk liquid phase and the value of reaction rate is calculated under the bulk liquid phase conditions in reactor steady state. The value of the mass transfer coefficient $k_{\ell,k}$ was obtained using correlations for mechanically stirred G–L tanks ([13] and [8]), and the same

Table 3
Parameters of the feeds

Case		T (K)	Phase	$\dot{F}$(mol s^{-1})	x_A	x_B	x_R	x_S
Poor gas	1	330.15	Liquid	1.944	0.7	–	–	0.3
	2	352.15	Gaseous	1.389	–	0.2	–	0.8
Rich gas	1	330.15	Liquid	0.833	0.6	–	–	0.4
	2	352.15	Gaseous	1.389	–	0.98	–	0.02

value has been set for all other compounds (A, R, S). Thus, to vary conditions in the reactor to obtain a different Hatta number, only the value of the reaction rate constant has been changed in wide range. Very small values of the Hatta number mean a slow reaction and a practically negligible mass transfer resistance. In such case the concentration profiles in the liquid film are very flat, overall consumptions of reactants in the film are negligible compared to those in the bulk liquid phase, and the reactor can be simply described by the equilibrium model. High values of the Hatta number mean a very fast chemical reaction and a high mass transfer resistance of the liquid film. The concentration profiles in the liquid film are very steep and description of the reactor performance using equilibrium model can provide unrealistic results, if any. On the other hand, when solving the reaction–diffusion equation (Eq. (1) with BC given by Eq. (2)) numerical problems can arise.

Two case studies have been solved, as is indicated in Table 3. In both cases reactant A has been fed into the reactor in liquid phase (with component S), reactant B has been fed into the reactor in gaseous phase (again with component S). In the first case (poor gas), reactant B has been fed at low concentration, and the ratio of its molar flow fed into the reactor and the molar flow rate of reactant A is 0.204. In the second case (rich gas), reactant B has been fed as quasi pure (98%), and the ratio of its molar flow fed into the reactor and molar flow rate of reactant A was 2.72.

The influence of the reaction rate constant on the Hatta number in both cases assumed above is depicted in Fig. 3. Because of high concentration of the key component B in the gaseous stream in Case 2, the driving force for its mass transfer from gaseous to liquid phase was very high and the reaction proceeds predominantly in the bulk liquid phase with high conversion of component A. The value of the Hatta number is in wide range of the reaction rate constants small (approximately 0.07). The depletion of the key component B in the liquid film is negligible (cf. Fig. 4). Because of moderate profiles of reactants A and B in the liquid film, the numerical algorithm for film model equations used only 30 discretization points for their discretization (the initial grid consisted of 20 points). The opposite situation occurs in Case 1—poor gas. Due to low amount of the key component B fed in the vapour phase into the reactor, the increase of the reaction rate constant has positive influence on the Hatta number increase (see Fig. 3). From the numerical point of view, to increase values of the Hatta number the number of discretization points in the liquid film increased rapidly, from about 20 for Hatta number values close to zero till 200 grid points for Hatta number equal 10. In Fig. 5 the concentration profiles of reactants in the liquid film are shown for $Ha = 10$. Concentration of the

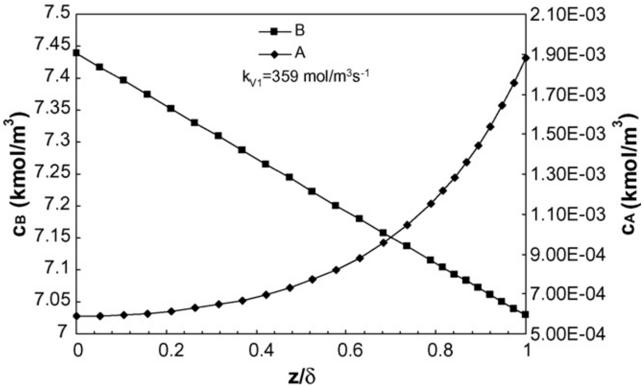

Fig. 4. Concentration of reactants in the liquid film for the Case 2—rich gas.

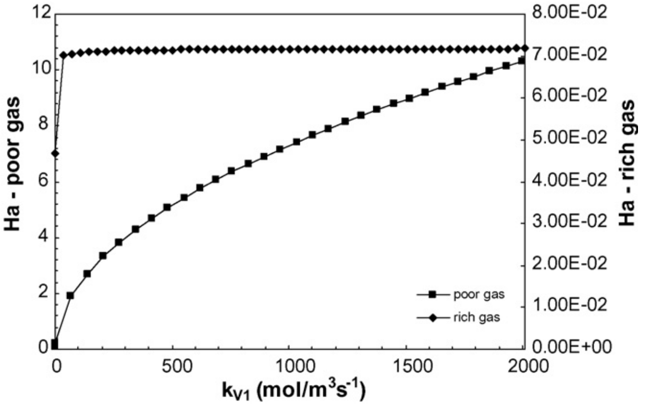

Fig. 3. Influence of the reaction rate constant on the Hatta number values in a CSTR with separation.

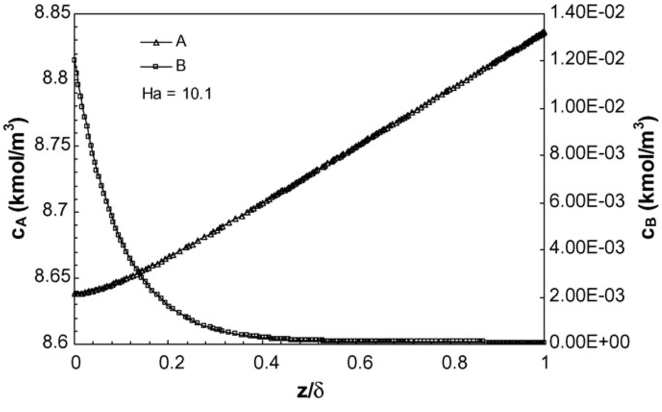

Fig. 5. Concentration profiles of the reactants A and B in the liquid film, Case 1—poor gas, $Ha = 10$.

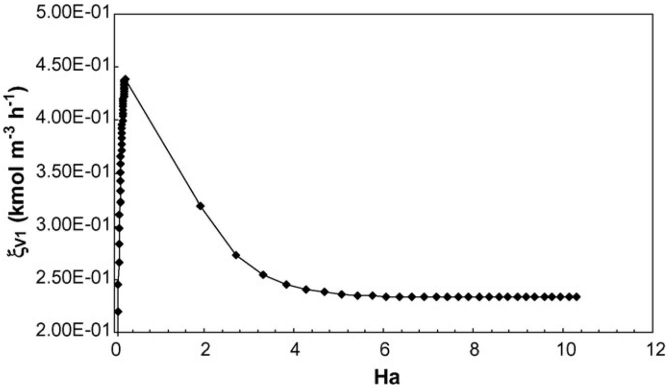

Fig. 6. Reaction rate in the reactor bulk liquid phase as a function of the Hatta number.

key component B in the liquid film decreased practically to zero in the first half of the film, e.g. almost all key component entering into the liquid phase is consumed in the liquid film, and its molar flux entering the liquid bulk is very small. Consequently the rate of chemical reaction in the bulk phase is very small. In Fig. 6, the dependence of the reaction rate in bulk phase on the Hatta number is demonstrated. For small values of the reaction rate constant (*Ha* less than 1) reaction is very slow, almost no reactant B is consumed in the liquid film and it reacts in the bulk liquid. Increasing the reaction rate constant has positive influence on the reaction rate in the bulk phase. A further increase of the reaction rate constant leads to a higher consumption of reactant B in the liquid film and to a decreased molar flow into the bulk liquid resulting in the decrease of the reaction rate.

5. Conclusion

The main objective of the presented paper was the analysis of a CSTR with reactive separation for fast homogeneous chemical reaction in the liquid phase. To describe the vapour–liquid interface, the film model has been applied. For fast reactions, the liquid film can play an important role and can significantly influence the reactor behaviour. Mathematical model of steady-state CSTR coupled by the reaction–diffusion equations describing the vapour–liquid interface is presented accompanied by the algorithm of the model equations solution. The model equations which are strongly non-linear in combination with the steep concentration profiles in the liquid film caused by a fast reaction can lead to numerical problems. To avoid this, the bulk phase material balances and the liquid film equations were treated separately, as is described in Fig. 2. To solve the reaction–diffusion equations, the non-uniform discretization procedure for space variable from the IMSL Math library [11] has been used. Model reaction system involving four components and one reversible reaction has been chosen. For simulations purposes, the physical and chemical properties as well as the V–L equilibria data for acetic acid esterification by ethanol have been used. The algorithm has been tested for different values of the Hatta number until values above 10 when the concentration profile of the key component was very steep and its molar flux entering the bulk liquid was very low. The proposed algorithm is very robust and straightforward even for badly chosen starting values of variables, both in bulk phases and in the liquid film. In the future, the presented algorithm will be extended and used for reactive distillation column simulation, and the results will be compared with the MS approach as described by Frank et al. [3].

We would like to point out that the proposed algorithm cannot be used for instantaneous reactions with “infinitely large” Hatta numbers. For such cases the approximate solutions proposed by Astarita [14], Danckwerts [15] can be used.

Acknowledgements

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