

Supplementary Materials: Model-Based Investigation of the Interaction of Gas-Consuming Reactions and Internal Circulation Flow Within Jet Loop Reactors

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S1. Parameter applied in the simulation studies

S1.1. Constant parameters

Table S.1 summarizes constant parameters applied in the simulations: property data, reactor geometry and process conditions.

Table S.1. Left: Property data applied in the simulations. Center: Constant reactor geometry parameter. Right: Constant process conditions.

Property data			Reactor geometry			Process conditions		
Parameter	Unit	Value	Parameter	Unit	Value	Parameter	Unit	Value
ρ_l	kg m^{-3}	1000	V_R	m^3	3.12	f_s	-	1.1
ρ_g	kg m^{-3}	8.17	V_{ER}	m^3	0.2	p_0	bar	11.11
M_A	kg kmol^{-1}	40	ζ_N	-	1.08	$V_{l,\text{feed}}$	$\text{m}^3 \text{s}^{-1}$	$1 \cdot 10^{-4}$
M_B	kg kmol^{-1}	28	d_s	m	0.002			
$\nu_{A,I}$	-	-1	h_{BS}	m	1.50			
$\nu_{B,I}$	-	-1	h_{HS}	m	0.80			
ν_l	$\text{m}^2 \text{s}^{-1}$	$1 \cdot 10^{-6}$	h_{BR}	m	0.25			
			h_{HR}	m	0.35			

Where, ν_l is the kinematic viscosity of the liquid phase and d_s is the wall thickness of the draft tube.

S1.2. Parameters study I

Table S.2 summarizes the applied parameters of study I.

Table S.2. Applied parameters of study I.

Parameter	Unit	Values/Range
f_r	-	1.0
k_I	s^{-1}	[0,2]
φ	kW m^{-3}	(1.13,1.70,4.00)
d_N	cm	2.5
d_R	m	0.70
d_{DT}	m	0.35
h_R	m	8.1
h_{DT}	m	6.0
d_{DT}/d_R	-	0.5
h_R/d_R	-	11.6

S1.3. Parameters study II

Table S.3 summarizes the applied parameters of study II where the diameter ratio is varied.

Table S.3. Applied parameters of study II where the diameter ratio is varied.

Parameter	Unit	Values/Range
f_r	-	1.0
k_I	s^{-1}	(0.0,0.2,2.0)
φ	$kW m^{-3}$	(1.13,1.7,4.0)
V_{ER}	$m^3 s^{-1}$	0.0177
d_R	m	0.7
d_{DT}	m	[0.12,0.37]
h_R	m	8.1
h_{DT}	m	6.0
d_{DT}/d_R	-	[0.175,0.525]
h_R/d_R	-	11.6

Table S.4 summarizes the applied parameters of study II where the slenderness ratio is varied.

Table S.4. Applied parameters of study II where the slenderness ratio is varied.

Parameter	Unit	Values/Range
f_r	-	1.0
k_I	s^{-1}	(0.0,0.2,2.0)
φ	$kW m^{-3}$	(1.13,1.7,4.0)
V_{ER}	$m^3 s^{-1}$	0.0177
d_R	m	Equation (S.1)
d_{DT}	m	Equation (S.2)
h_R	m	Equation (S.3)
h_{DT}	m	Equation (S.4)
d_{DT}/d_R	-	0.5
h_R/d_R	-	[7,15]

To keep the reactor volume constant, the Equations (S.1)-(S.4) are applied.

$$d_R = \left(\frac{V_R}{\frac{\pi}{4} \frac{h_R}{d_R}} \right)^{1/3} \quad (S.1)$$

$$d_{DT} = \frac{d_{DT}}{d_R} d_R \quad (S.2)$$

$$h_R = \frac{h_R}{d_R} d_R \quad (S.3)$$

$$h_{DT} = h_R - h_{BR} - h_{BS} - h_{HS} \quad (S.4)$$

S1.4. Parameters study III

Table S.5 summarizes the applied parameters of study III.

Table S.5. Applied parameters of study III.

Parameter	Unit	Values/Range
f_r	-	(0, 7]
k_I	s^{-1}	(0,0.2,2)
φ	$kW m^{-3}$	(1.13,1.7,4.0)
V_{ER}	$m^3 s^{-1}$	0.0177
d_R	m	0.7
d_{DT}	m	(0.21,0.35)
h_R	m	8.1
h_{DT}	m	6.0
d_{DT}/d_R	-	(0.3,0.5)
h_R/d_R	-	11.6

S2. Modeling details

S2.1. Pressure loss coefficient

The pressure loss coefficient ζ within compartments DT and AG are calculated according to Idel chik [1] with:

$$\zeta_i = \lambda \frac{z}{d_i} \quad \text{for } i \in \{DT, AG\}, \quad (S.5)$$

where λ is the Darcy friction factor, which is computed using Equation (S.6).

$$\lambda = \begin{cases} 64/Re & Re \leq 3200 \\ 0.03 & Re > 3200 \end{cases} \quad (S.6)$$

Here, Re is the Reynolds number which is defined by:

$$Re_i = \frac{u_i d_i}{\nu_l} \quad \text{for } i \in \{DT, AG\}. \quad (S.7)$$

For compartment DT, the pipe diameter d_{DT} is equal to the hydraulic diameter because it is a cylindrical pipe. For compartment AG, the hydraulic diameter of the circular section is calculated according to Equation (S.8).

$$d_{AG} = d_R - d_{DT} - 2 d_s \quad (S.8)$$

An additional pressure loss occurs during the transition from compartment DT to AG and vice versa due to the flow redirection. The pressure loss coefficient in JLR was intensively investigated by Zehner [2] and resulted into the applied formulas:

$$\zeta_{DT \rightarrow AG} = 1.8 a_{DT} + 3.0 \left(\frac{a_{DT}}{a_{AG}} \right) \exp \left[-100 \frac{d_s}{d_R} \right], \quad (S.9)$$

$$\zeta_{AG \rightarrow DT} = 3.6 a_{DT} + 1.5 \left(\frac{a_{DT}}{a_{AG}} \right) \exp \left[-50 \frac{d_s}{d_R} \right], \quad (S.10)$$

where, a_i is the ratio of the cross-sectional area of the compartment i to the cross-sectional area of the reactor (see Equation (S.11)) and d_R is the diameter of the reactor.

$$a_i = \frac{A_i}{A_R} \quad \text{for } i \in \{DT, AG\} \quad (S.11)$$

Here, A_R is the cross-sectional area of the reactor.

S2.2. Ideal reactor models

In following sections the plug flow reactor model (PFR) and the continuous stirred tank reactor (CSTR) model for the calculation of the concentrations present in the compartments are presented. A detailed derivation of the subsequent equations can be found in Octave Levenspiel [3].

S2.2.1. Plug flow reactor model

Based on the plug flow model, the Equation (S.12) describing the propagation of the concentration of species A can be derived under additional assumptions: isothermal conditions (k_1 is constant), single, irreversible reaction of first-order.

$$c_{l,A,i} = c_{l,A,IN,i} \exp(-Da_i) \quad (S.12)$$

Here $c_{l,A,IN,i}$ is the molar concentration of species A at the respective inlet of the compartment i , Da_i is the first-order Damköhler number of the compartment i . Applying the Equation (S.12) to the compartments results in the following equations. Thereby it is assumed that the concentration at the outlet of compartment DT is the same as the inlet of compartment AG.

$$c_{l,A,DT} = c_{l,A,DT,IN} \exp(-Da_{DT}) \quad (S.13)$$

$$c_{l,A,AG} = c_{l,A,DT,OUT} \exp(-Da_{AG}) \quad (S.14)$$

$$c_{l,A,ER} = c_{l,A,BS} \exp(-Da_{ER}) \quad (S.15)$$

with

$$c_{l,A,AG,IN} = c_{l,A,DT,OUT} \quad (S.16)$$

S2.2.2. Continuous stirred tank reactor model

Based on the continuous stirred tank reactor model, the Equation (S.17) describing the molar concentration of species A in the liquid phase can be derived by assuming the same additional assumptions as in Section S2.2.1.

$$c_{l,A,i} = c_{l,A,IN,i} \frac{1}{1 + Da_i} \quad (S.17)$$

If the Equation (S.17) is applied to the compartment BS, it follows:

$$c_{l,A,BS} = c_{l,A,DT,OUT} \frac{1}{1 + Da_{BS}} \quad (S.18)$$

S2.3. Inlet conditions draft tube

S2.3.1. Concentration of species A

To calculate the concentration at the inlet of the draft tube, it is assumed that the incoming flows ($\dot{V}_{l,feed}$, $\dot{V}_{l,AG}$, \dot{V}_{ER}) are instantaneously mixed with each other and no reaction takes place. From the mass and species balance it follows:

$$c_{l,A,DT,IN} = \frac{\dot{V}_{l,feed} \frac{\rho_l}{M_A}}{\dot{V}_{l,DT,IN} - \dot{V}_{l,AG} \exp(-(\overline{Da}_{AG} + \overline{Da}_{DT})) - \dot{V}_{ER} \frac{\exp(-\overline{Da}_{DT})}{1 + Da_{BS}}} \quad (S.19)$$

Here, \overline{Da}_i is the integral Damköhler number of the compartment i .

S2.3.2. Velocity of the liquid phase

To determine the velocity of the liquid phase at the inlet of the draft tube, a momentum flow balance is drawn around an infinitely small control volume (see Equation (S.20)). Where the momentum flow of the liquid feed was neglected.

$$\dot{V}_{l,DT,IN} u_{l,DT,IN} = \dot{V}_{ER} u_{l,ER} + \dot{V}_{l,AG} u_{l,AG,DT,IN} - \frac{\zeta_{AG \rightarrow DT}}{2} A_{DT} u_{l,DT,IN}^2 \quad (S.20)$$

Here $u_{l,AG,DT,IN}$ is a fictive velocity of the liquid phase of the volume flow coming from the annular gap into the draft tube. This volume flow is not related to the entire cross-sectional area of the draft tube, but to the annulus created by the nozzle mouth area. The Equation (S.21) is used to calculate this fictive velocity

$$u_{l,AG,DT,IN} = \frac{u_{l,AG,OUT} A_{AG}}{A_{DT,IN}}, \quad (S.21)$$

with

$$A_{DT,IN} = A_{DT} - A_N. \quad (S.22)$$

S2.4. Damköhler number

The first-order Damköhler number describes the ratio of the rate constant of the reaction to the rate constant of the convective mass transport. It could be shown that

$$Da_i(z) = k_1 \tau_i(z), \quad (S.23)$$

is valid. Here, $\tau(z)$ is the residence time in the compartment i . The integral Damköhler number of the compartment i is calculated as follow:

$$\overline{Da}_i = \frac{1}{h_i} \int_0^{h_i} Da_i(z) dz. \quad (S.24)$$

S2.5. Residence behavior

The residence time in compartment i describes the ratio between reaction volume and volume flow rate through the reaction volume. The volume flow rate of the liquid phase in the different compartments are calculated as follows. In addition the equations for $\dot{V}_{l,product}$ and $\dot{V}_{g,purge}$ are given.

$$\dot{V}_{l,DT,IN} = \dot{V}_{l,N} + \dot{V}_{l,feed} + \dot{V}_{l,AG} \quad (S.25)$$

$$\dot{V}_{l,DT} = \dot{V}_{l,DT,IN} \quad (S.26)$$

$$\dot{V}_{l,BS} = \dot{V}_{l,N} + \dot{V}_{l,feed} \quad (S.27)$$

$$\dot{V}_{l,product} = \dot{V}_{l,feed} \quad (S.28)$$

$$\dot{V}_{g,purge} = \dot{V}_{g,feed} (f_s - 1) \quad (S.29)$$

Using this the residence time in the different compartments results in

$$\tau_{\text{DT}}(z) = \alpha_{\text{l,DT}}(z) \frac{V_{\text{DT}}(z)}{\dot{V}_{\text{l,DT}}}, \quad (\text{S.30})$$

$$\tau_{\text{BS}} = \frac{V_{\text{BS}}}{\dot{V}_{\text{BS}}}, \quad (\text{S.31})$$

$$\tau_{\text{AG}}(z) = \alpha_{\text{l,AG}}(z) \frac{V_{\text{AG}}(z)}{\dot{V}_{\text{l,AG}}}, \quad (\text{S.32})$$

$$\tau_{\text{ER}} = \frac{V_{\text{ER}}}{\dot{V}_{\text{l,ER}}}. \quad (\text{S.33})$$

Where V_i is the reaction volume in compartment i , which are summarized in Equation (S.34) to (S.36).

$$V_{\text{DT}}(z) = A_{\text{DT}} z \quad (\text{S.34})$$

$$V_{\text{BS}} = A_{\text{BS}} h_{\text{BS}} \quad (\text{S.35})$$

$$V_{\text{AG}}(z) = A_{\text{AG}} z \quad (\text{S.36})$$

The occurring cross-sectional areas A_i in Equations (S.22), (S.34)-(S.36) are calculated according to the following equations:

$$A_{\text{R}} = \frac{\pi}{4} d_{\text{R}}^2, \quad (\text{S.37})$$

$$A_{\text{N}} = \frac{\pi}{4} d_{\text{N}}^2, \quad (\text{S.38})$$

$$A_{\text{DT}} = \frac{\pi}{4} d_{\text{DT}}^2, \quad (\text{S.39})$$

$$A_{\text{BS}} = A_{\text{R}}, \quad (\text{S.40})$$

$$A_{\text{AG}} = A_{\text{R}} - A_{\text{DT}} - \pi d_{\text{DT}} d_{\text{s}}. \quad (\text{S.41})$$

S3. Additional figures

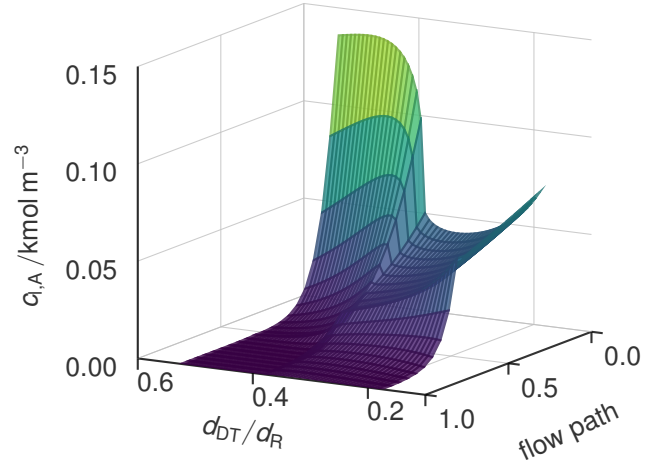


Figure S.1. Molar concentration of species A in the liquid phase $c_{l,A}$ as a function of the diameter ratio d_{DT}/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.3 with $k_1 = 0.2 \text{ s}^{-1}$ where the reactor volume where kept constant.

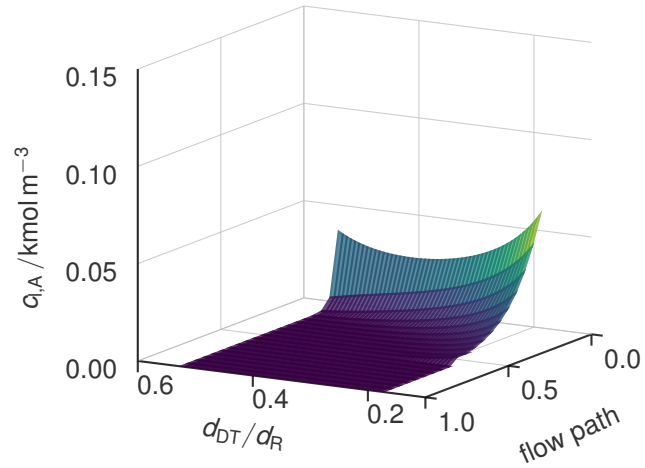


Figure S.2. Molar concentration of species A in the liquid phase $c_{l,A}$ as a function of the diameter ratio d_{DT}/d_R and the normalized flow path using the parameters specified Table S.1 and Table S.3 with $k_1 = 2.0 \text{ s}^{-1}$ where the reactor volume where kept constant.

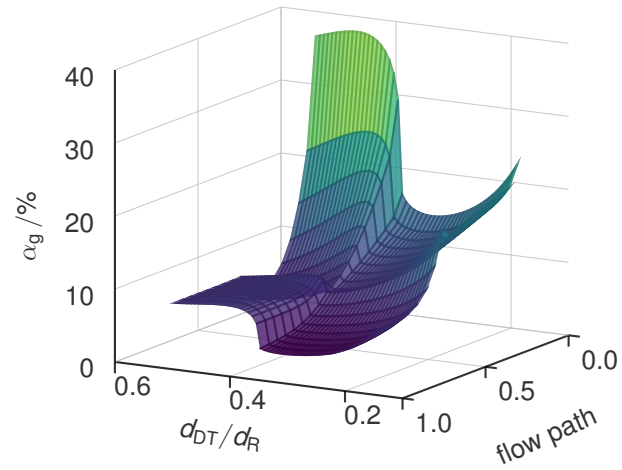


Figure S.3. Gas volume fraction α_g as a function of the diameter ratio d_{DT}/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.3 with $k_l = 0.2 \text{ s}^{-1}$ where the reactor volume where kept constant.

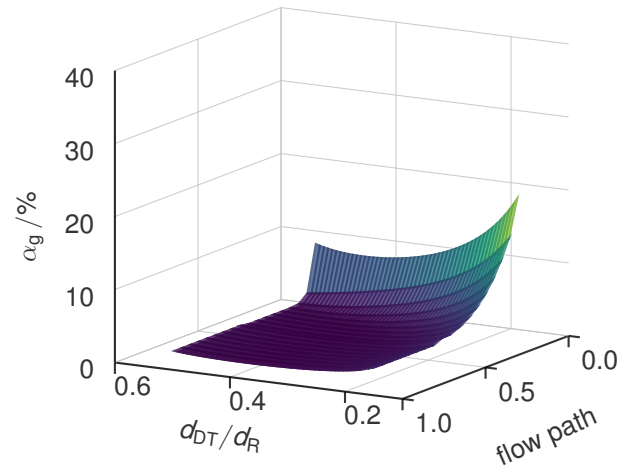


Figure S.4. Gas volume fraction α_g as a function of the diameter ratio d_{DT}/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.3 with $k_l = 2.0 \text{ s}^{-1}$ where the reactor volume where kept constant.

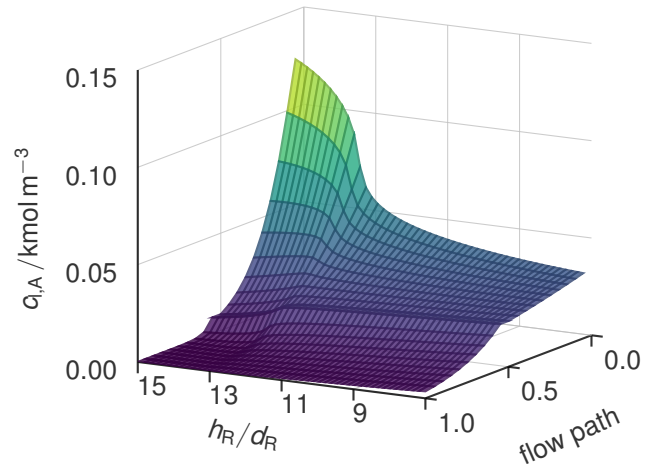


Figure S.5. Molar concentration of A in the liquid phase $c_{l,A}$ as a function of the slenderness ratio h_R/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.4 with $k_l = 0.2 \text{ s}^{-1}$ whereby the diameter ratio $d_{DT}/d_R = 0.5$ and the reactor volume were kept constant.

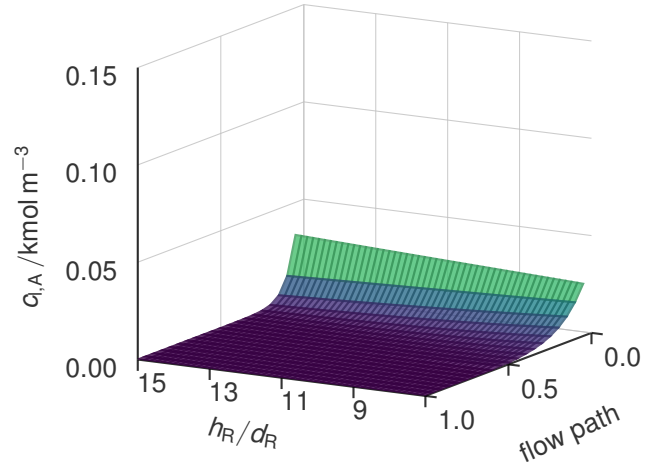


Figure S.6. Molar concentration of A in the liquid phase $c_{l,A}$ as a function of the slenderness ratio h_R/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.4 with $k_l = 2.0 \text{ s}^{-1}$ whereby the diameter ratio $d_{DT}/d_R = 0.5$ and the reactor volume were kept constant.

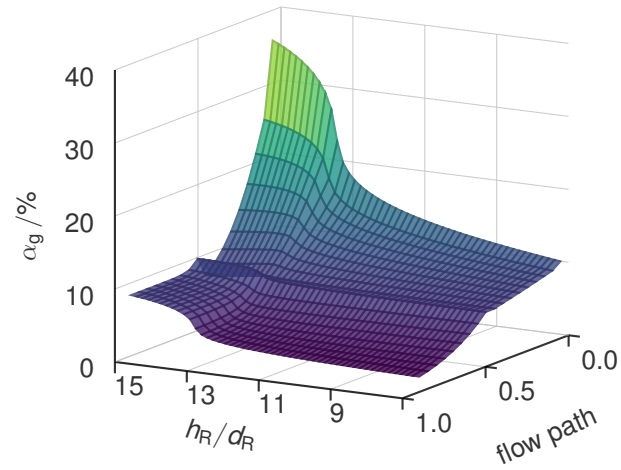


Figure S.7. Gas volume fraction α_g as a function of the slenderness ratio h_R/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.4 with $k_1 = 0.2 \text{ s}^{-1}$ whereby the diameter ratio $d_{DT}/d_R = 0.5$ and the reactor volume were kept constant.

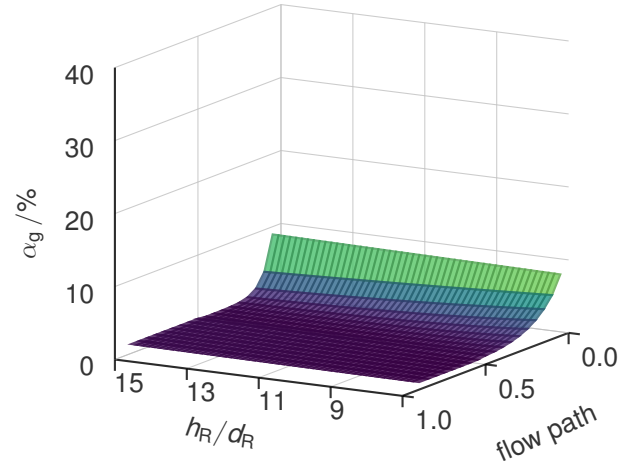


Figure S.8. Gas volume fraction α_g as a function of the slenderness ratio h_R/d_R and the normalized flow path using the parameters specified in Table S.1 and Table S.4 with $k_1 = 2.0 \text{ s}^{-1}$ whereby the diameter ratio $d_{DT}/d_R = 0.5$ and the reactor volume were kept constant.

S4. Implementation details

Figure S.9 shows a flowchart of the solution procedure. On average, a single simulation takes less than a minute on an office notebook (CPU: AMD Ryzen 7 PRO4750 U with max. 4.1 GHz; RAM: 16 GB with 3200 MHz).

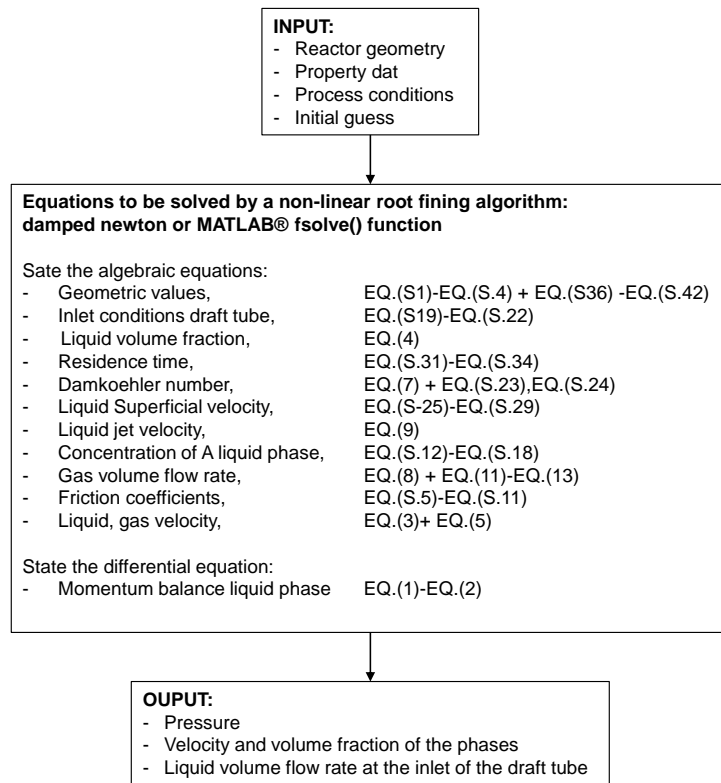


Figure S.9. Flowchart of the solution procedure.

Nomenclature

The following nomenclature is used in this manuscript:

Latin Letters

a_i	Ratio of the cross-sectional area of compartment i to the cross-sectional area of the reactor	
A_i	Cross-sectional area of compartment i	m^2
$c_{\text{LA},i}$	Molar concentration of species A in the liquid phase in compartment i	mol m^{-3}
$c_{\text{LA,IN},i}$	molar concentration of species A at the respective inlet of the compartment i	mol m^{-3}
d_i	Hydraulic diameter of compartment i	m
Da_i	Damköhler number of first order of compartment i	
\overline{Da}_i	Integral Damköhler number of first order of compartment i	
f_r	Recycling excess factor	
f_s	Stoichiometric excess factor of species B	
F_l	Friction force per unit volume acting on the liquid phase	N m^{-3}
g	Gravitational acceleration constant	m s^{-2}
h_i	Height of compartment i	m
k_{I}	Rate of reaction constant of Reaction I	s^{-1}
M_j	Molar mass of species j	mol kg^{-1}
N_{circ}	Circulation number	
p_0	Pressure in the headspace	bar
Δp_{N}	Pressure drop over the nozzle	Pa
p	Pressure	Pa
P	Power introduced into the reactor	W
r_{I}	Rate of reaction per unit liquid volume of Reaction I	$\text{mol m}^{-3} \text{s}^{-1}$
Re_i	Reynolds number of compartment i	
u_k	Velocity of phase k	kg m^{-3}
$u_{\text{LAG,DT,IN}}$	Fictive velocity of the liquid phase of the volume flow coming from the annular gap into the draft tube	kg m^{-3}
$\dot{V}_{k,i}$	Volume flow rate of phase k in compartment i	$\text{m}^3 \text{s}^{-1}$
V_i	Volume of compartment i	m^3
z	Coordinate along the axial direction	m
$z_{\text{IN},i}$	Initial position in flow path direction of compartment i	m

Greek letters

α_k	Volume fraction of phase k	
$\Delta\bar{\alpha}_{g,DT,AG}$	Difference between the gas hold up of compartment DT and AG; gas holdup difference	
$\bar{\alpha}_{k,i}$	Gas hold up of phase k of compartment i , integral over $\alpha_{k,i}(z)$ along z	
ζ	Pressure loss coefficient	
λ	Darcy friction factor	
ν_l	Kinematic viscosity of the liquid phase	$\text{m}^2 \text{s}^{-1}$
$\nu_{l,j}$	Stoichiometric coefficient of chemical species j of the reaction I	
ρ_k	Mass density of phase k	kgm^{-3}
τ_i	Residence time in compartment i	s
φ	Specific power input per reactor volume; power input	W m^{-3}

Subscripts

circ	Circulation
feed	Feed stream
g	Gas phase index
i	Compartment index
IN	Inlet
I	Reaction I
j	Chemical species index
k	Phase index
l	Liquid phase index
OUT	Outlet
product	Product stream
purge	Purge stream
s	Quantity related to the wall of the draft tube

Abbreviations

A	High-boiling reactant A
AH	Aerate height
AG	Annular gap
B	Chemical species B
BP	Baffle plate
BR	Bottom redirection zone
BS	Bottom space
C	High-boiling product C
CFD	Computational fluid dynamics
CSTR	Continuous stirred tank reactor
DT	Draft tube
ER	External recycle
HR	Head redirection zone
HS	Head space
JLR	Jet loop reactor
N	Nozzle
PBE	Population balance equation
PFR	Plug flow reactor
R	Reactor

References

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