Modeling and simulation of an industrial three phase trickle bed reactor responsible for the hydrogenation of 1,3-butadiene: A case study

T. Tohidian [a], O. Dehghani [a], M.R. Rahimpour [a,b],*

[a] Department of Chemical Engineering, School of Chemical and Petroleum Engineering, Shiraz University, Shiraz 71345, Iran
[b] Department of Chemical Engineering and Materials Science, University of California, 1 Shields Avenue, Davis, CA 95616, United States

HIGHLIGHTS

- Modeling of an industrial trickle bed reactor responsible for the hydrogenation of 1,3-butadiene into n-butane.
- Developing a more detailed reaction network.
- Investigation of the effect of inlet temperature on the behavior of the reactor.
- Studying the performance of the reactor under different operating conditions of the flowing feed stream.

ARTICLE INFO

ABSTRACT

In this research, an industrial trickle bed reactor responsible for the hydrogenation of 1,3-butadiene into n-butane has been chosen as a case study. In this regards, a suitable reaction network has been applied as the base kinetic structure and then it has been developed further to a more detailed reaction scheme capable of predicting available plant data. As the next step, attempts have been made to establish an accurate and simple to use mathematical modeling with the ultimate goal of predicting the plant outputs. The results ascertained the success of the proposed modeling in terms of total relative error of about 0.1. Moreover, the behavior of different parameters including temperature and molar flow rates along the length of the reactor has been studied. Additionally, the effect of inlet temperature on the behavior of the understudied trickle bed reactor has been seriously investigated. Finally, the performance of the three phase catalytic reactor has been studied under different operating conditions of the flowing feed stream.

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1. Introduction

Catalytic trickle bed reactors (TBRs) are among multiphase systems wherein a fixed bed of catalyst particles is in concurrent contact with a gas–liquid stream flowing downward the reactor [1–5]. Their application areas encompass a broad spectrum of industries from chemical, petrochemical, and petroleum to waste water treatment, biochemical, pharmaceutical, and electrochemical industries [3–8]. They are advantageously applied for heterogeneously

catalyzed reactions including hydrogenation, oxidation, alkylation, and chlorination [1,9–14]. TBRs have superiority over other three-phase catalytic reactors in various aspects. Since it is impossible to enumerate here all advantages of such reactors, only some outstanding features are listed below [3,4,10,11,15]:

- Simplicity of operation due to no moving parts.
- Lower catalyst attrition due to their simple construction.
- Larger reactor size.
- Lower liquid holdup that further results in lower homogeneous side-reactions.
- Lower pressure drop.
- High conversion due to plug flow pattern of the liquid phase.
- Lower energy dissipation rate.

In this study, advantages of TBRs are applied in order to purify a C_4 cut stream existed in an olefin complex. Generally, C_4 cut refining cannot be carried out practically by physical separation

Nomenclature

A_c	cross sectional area of the reactor (m^2)
C	molar concentration (mol m^3)
C_{pi}°	standard heat capacity of ith component at the standard temperature (J mol^{-1} k^{-1})
d_p	particle diameter (m)
d_r	reactor diameter (m)
D_{l,H_2}	hydrogen diffusivity in liquid phase (m^2 s^{-1})
E	activation energy (J mol^{-1})
F	molar flow rate (mol s^{-1})
G_i°	Gibbs energy of ith component at the standard state pressure
G_{0i}°	standard Gibbs energy of formation of ith component at the standard temperature (J mol^{-1})
h	Henry's constant (pa m^3 mol^{-1})
H_{0i}°	standard enthalpy of formation of ith component at the standard temperature (J mol^{-1})
k	frequency factor (refer to Table 8 for units)
$(ka)_{gl}$	gas–liquid volumetric mass transfer coefficient (s^{-1})
$(ka)_{ls}$	liquid–solid volumetric mass transfer coefficient (s^{-1})
K_{eq}	equilibrium constant
k_{ref}	frequency factor at reference temperature (refer to Table 8 for units)
M	molecular weight (gr.mol^{-1})
P_r	reduced pressure
R	universal gas constant (J mol^{-1} k^{-1})
Re	Reynolds number
r_j	rate of reaction (mol s^{-1} kg cat^{-1})
Sc	schmidt number
s_p	shape factor
T	temperature (K)
T_r	reduced temperature
T_{ref}	reference temperature (K)

T_0	the standard temperature (298.15 K)
u	superficial velocity
V_{H_2}	molar gas volume of H_2 at standard conditions (m^3 - mol^{-1})
Z	differential length of the reactor (m)

Greek letter

ΔG°	Gibbs energy change when each species are at the standard state pressure
ΔH_{r_j}	heat of jth reaction (J mol^{-1})
ΔH_v	heat of vaporization (J mol^{-1})
ε_l	liquid holdup
v_i	stoichiometric number of ith component
ρ_B	density of catalytic bed (kg cat m^3)
μ	viscosity (kg m^{-1} s^{-1})
λ_{H_2}	solubility of hydrogen in hydrocarbon mixtures (m^3 - kg^{-1} pa^{-1})
ρ_l	liquid phase density at operating conditions (kg m^{-3})
ρ_{20}	liquid phase density at 20 °C (kg m^{-3})
φ	association parameter
υ	molar volume at normal boiling point (m^3 mol^{-1})

Subscript and superscript

i	counter of components
Ig	ideal gas
J	counter of reactions
G	gas
L	liquid
S	solid
T	total

processes such as distillation owing to small relative volatility of all species in the mixture. Industrially, unsaturated compounds of C_4 cut including mainly 1,3-butadiene (BD), 1-butene (1BE), cis 2-butene (cBE), trans 2-butene (tBE), and isobutane (IBA) can be purified by selective hydrogenation in TBRs to form more valuable saturated components such as n-butane (BA) and isobutane (IBA) [16–19]. Butane compounds (including both BA and IBA) with various ratios of propane are known as liquefied petroleum gas (LPG) that widely used for internal combustion engines as an energy-economy-ecology friendly fuel [20–22]. LPG with no ozone depletion potential can also be applied as an alternative to conventional chlorofluorocarbon refrigerants [20]. Moreover, BA can be applied in solid oxide fuel cell (SOFC) systems to produce low cost-low emission electricity [23].

1.1. A literature review on reactor modeling

In 2003, Bressa et al. conducted an experimental study to evaluate a kinetic expression for the catalytic hydrogenation of unsaturated traces in C_3–C_4 streams. Finally, they applied the results for 1-dimentional mathematical modeling of an adiabatic three-phase fixed-bed catalytic unit operated in up-flow mode with the hypotheses of complete wetting and no axial dispersion. The results introduced the temperature rise as a detrimental factor for the reactor performance. However, the simulation analysis attributed the higher performance of the reactor to both H_2 input load enhancement and the operating pressure rise [24]. In the same year, the research group of Dietz et al. developed a steady state model of a trickle bed reactor for the consecutive

hydrogenation of 1,5,9-cyclododecatriene. They proposed a non-isothermal heterogeneous model taking into account the partial wetting of the catalyst, as well as the resistances to heat and mass transfer at the gas–liquid, liquid–solid and solid–gas interfaces [25].

Shortly after, Lange and colleagues conducted an experimental and theoretical study of the hydrogenation of alpha-methylstyrene in a laboratory-scale trickle-bed reactor under forced periodic operation. In this regards, they made use of an extended axial dispersion model incorporating the variation of partially wetted catalyst surface. The results showed higher time-average conversion of alpha-methylstyrene in the forced operation mode in comparison to the steady state operating condition. The authors attributed the result to the variation in liquid hold-up and wetting of the catalyst particles [1].

In 2008, Liu and coworkers selected the hydrogenation reaction of dicylcopentadiene in a trickle-bed reactor as a case study. They investigated the effect of five operation strategies including ON–OFF and PEAK–BASE modulations of the liquid flow rate or concentrations respectively, and a novel hybrid modulation of both liquid flow rate and concentration. Finally, it was revealed that the modulation of liquid flow rate improve the hydrogenation reaction [26].

In 2012, Iliuta and Iliuta performed a two-dimensional, non-isothermal, unsteady-state mathematical modeling that considered momentum, mass, and enthalpy transport processes in the case of sulfur removal from gasoil by applying a monolith three-phase reactor. They compared the reactor performance with that of a conventional trickle bed reactor operated at the same liquid