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Optimization of process-specific catalytic packing in catalytic distillation process: A multi-scale strategy

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HIGHLIGHTS

- A microscopic model focused on reactive performance was established.
- A two-way coupling multi-scale model was proposed for catalytic distillation.
- The simulation results were in good agreement with experimental data.
- The effect of equivalent diameter of catalyst layer on efficiency factor was studied.

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ABSTRACT

A novel two-way coupling multi-scale model was proposed to investigate the catalytic distillation process. In the model, a microscopic model that focuses on the reactive performance of structure catalytic packing was used to calculate actual rate for catalytic distillation process, which is the basic parameter for process simulation. Furthermore, the traditional process simulation was used to provide proper boundary conditions for microscopic model. In order to validate the multi-scale model, heterogeneously catalyzed hydrolysis of methyl acetate was employed as a test system. The simulated final conversions of methyl acetate and catalyst layer efficiency factors were in good agreement with experimental results. The results indicated that as the equivalent diameter of catalyst layer decreases from 25.4 mm to 8.1 mm, the catalyst layer efficiency factor rises to 200% approximately. This study could provide a theoretical guide for the optimization of catalytic packing structure.

1. Introduction

Catalytic distillation (CD), which integrates the catalytic reaction and distillation in a single multifunctional process unit, is a well-known example of process intensification. Due to vast advantages such as high reaction conversion, low energy consumption, energy savings and simple operation, it has been successfully applied in esterification, etherification, hydrolyzation of ester, hydration of olefins, etc. (Harmsen, 2007). The type, geometry and structure of internals influence the whole process performance significantly in the catalytic distillation process. Thus, the developed catalytic packings have to enhance both separation and reaction, and maintain a sound balance between them. At present, a variety of structured catalytic packings have been developed (Subawalla et al., 1997; Götze et al., 2001; Odziej et al., 2005; Ding et al., 2015). However, these packings cannot be efficiently suitable for all the catalytic distillation process. Therefore, the development of a process-specific and high-separation-efficiently structured catalytic packings is desirable. Nevertheless, due to the complexity resulting from the nonideality of components and integrations among hydrodynamics, vapor-liquid mass transfer, inter diffusion and chemical kinetics, our cognition to the catalytic distillation process is insufficient. Until now, the design and optimization of structured catalytic packings mainly rely on engineering experience and empirical correlations.

In recent years, computational fluid dynamics (CFD) was widely used to investigate the local flow and mass transfer characteristics, and provided a new idea for the design of process-specific catalytic packing. For instance, the works by Higler et al. (1999), Van Baten et al. (2001), Van Baten and Krishna (2001, 2002), Dai et al. (2012), Li et al. (2012) and Ding et al. (2014). Nevertheless, influence of catalytic packing structure on macro variables such as conversion, yield and production purity which take many attentions in the actual industrial processes are unavailable simply based on CFD simulations. These macro variables can be obtained via process simulation which is widely described by equilibrium (EQ) and non-equilibrium (NEQ) models (Klöker et al., 2005; Zhang et al.,

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Nomenclature

A	surface area of catalyst particles inside the geometry domain, m^2
$A_{\gamma\kappa}$	interface of liquid phase and solid phase
$\mathbf{B}$	the matrix defined by Eq. (11)
C_{i0}	initial concentration of each component, $mol \cdot m^{-3}$
C_i	actual concentration inside the catalytic sheets, $mol \cdot m^{-3}$
D_{iE}	diffusion coefficient of each component, $m^2 \cdot s^{-1}$
$Đ_{AE}$	binary Maxwell-Stefan diffusion coefficient, $m^2 \cdot s^{-1}$
D	equivalent diameter of catalytic sheets, mm
d_p	catalyst particle diameter, mm
F	feed molar flow rate, $kmol \cdot h^{-1}$
K	chemical equilibrium constant
K_1	the constant defined by Eq. (17)
k_+,k_-	forward and backward reaction rate constant, $m^6 \cdot mol^{-1} \cdot s^{-1} \cdot (gcat)^{-1}$
L'	liquid molar flow rate at the feed stage, $kmol \cdot h^{-1}$
L	liquid reflux molar flow rate, $kmol \cdot h^{-1}$
m_{cat}	catalyst mass in one stage, g
M_i	molar mass, $kg \cdot kmol^{-1}$
$\mathbf{n}$	unit normal vector pointing from the fluid phase toward the solid phase
q	liquid fraction of feed
R_m	water to methyl acetate molar ratio
R_f	reflux rate to feed rate
R_i	chemical reactive source, $kg \cdot m^{-2} \cdot s^{-1}$
$r_{i-\ surface}$	reaction rate on the reactive surface, $mol \cdot m^{-2} \cdot s^{-1}$
r_{fact}	actual reaction rate of catalytic distillation process, $mol \cdot s^{-1}$
r_{ideal}	reaction rate in reactive kettle, $mol \cdot s^{-1}$
S_v	space velocity
T	reaction temperature, K
V	molar volume of solute at normal boiling point, $cm^3 \cdot mol^{-1}$
V_C	critical molar volume, $cm^3 \cdot mol^{-1}$
V_{bed}	volume of catalyst bales in one stage, m^3
V_{region}	volume of simulated geometry domain, m^3
$w_{E\gamma}$	mass fraction of each component in reaction system
w_{ib}	mass fraction of liquid component on the external surface of catalyst beds
W	concentration of catalyst pellets, $g \cdot m^{-3}$
$x_{i\gamma}$	mole fraction of each component
x_A	conversion of methyl acetate
Z_l	dynamic holdup, $m^3 \cdot m^{-3}$
Z_d	total holdup, $m^3 \cdot m^{-3}$

Greek letters

ε_γ	void fraction of porous media
ρ	mixture density, $kg \cdot m^{-3}$
μ^L	viscosity, cp
η	catalyst layer efficiency factor
ε_{BC}	volume ratio of catalytic sheets to catalyst bales
Γ_{AB}	thermodynamic factor defined by Eq. (14)
δ_{AB}	the constant defined by Eq. (15)

Abbreviations

CD	catalytic distillation
CFD	computational fluid dynamic
EQ	equilibrium
H2O	water
HAC	acetic acid
MeOAc	methyl acetate
MeOH	methanol
NEQ	non-equilibrium

2011). However, influence of structure on macro parameters cannot be considered directly during the process simulation. Therefore, it is impossible to realize optimization of process-specific structured catalytic packings simply based on either CFD simulation or process simulation.

Multi-scale model is an efficient method to establish the relationship between micro phenomenon and macro process (Sun et al., 2013). Liu et al. (2013) proposed a multi-scale model which combined CFD with process simulation to investigate the process of removal of acetic acid from water. Fluent software was used to calculate the tray efficiency in the element scale. And the calculated results were input into process simulation. With respect to the development of structured catalyst packing, Klöker et al. (2003), Górak et al. (2005) and Egorov et al. (2005) proposed a novel multi-scale model to obtain simulated results associated with hydraulic properties and mass transfer performance of different internals by using CFD approach, which were input into the rigorous, rate-based process simulation approach. Actually, not only the hydrodynamic and mass transfer characteristic of catalytic packing internal, but also the multicomponent mass transfer and reaction process in the catalyst layers influence the efficiency of catalytic distillation process. For the development of high-efficiency and process-specific catalytic packing, the determination of structural parameters of catalyst layers is also a key problem. To the best of our knowledge, none of the previous multi-scale models considered the influence of structural parameters of catalyst layers.

In the study, a novel two-way coupling multi-scale model that focuses on reactive performance of structured catalytic packing is proposed with the consideration of the inter influence between catalyst layer structure and catalytic distillation process. A microscopic model is used to investigate the multicomponent mass transfer and reaction process in the catalyst layer. The catalyst layer efficiency factor which is introduced to assess the rationality of catalyst layer structure parameter and obtained by experimental data or empirical method normally (Sundmacher and Hoffmann, 1993; Xu et al., 1995) can be calculated via the microscopic model. Then the actual reactive rate during catalytic distillation process which is the basic parameter for process simulation can be obtained. The traditional process simulation is used to provide proper boundary conditions for the microscopic model. A well-known reference system heterogeneously catalyzed hydrolysis of methyl acetate is employed as a test system, so as to validate the proposed novel multi-scale model. By the utilization of this proposed multi-scale model, the corresponding catalyst layer structural parameter can be determined for different engineering processes.

2. A multi-scale strategy

2.1. Multi-scale structures of catalytic distillation column

Catalytic distillation is characterized by hierarchical multi-scale structure in nature, as shown in Fig. 1, ranging from catalyst layer scale to the catalytic distillation column scale. This decomposition was based on the need of our study.

The catalyst layer scale is namely the microscopic scale in this work, which is generated by porous catalyst pellets packing. In this