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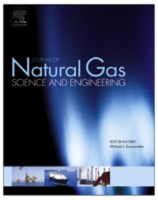

Effect of the flow pattern on catalytic reaction of propylene hydrogenation through structured catalyst bed: Mathematical modeling and experiment observations

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ABSTRACT

This novel study is experimental and modeling. In experimental section a pilot package including trickle bed reactor, automatic dosing pump, measuring instruments, gas chromatograph apparatus and five sample points along the reactor were employed to investigate the performance of propylene hydrogenation through structured catalyst bed under concurrent and countercurrent flow patterns. The experimental setup was configured in two different modes providing countercurrent and concurrent pattern for liquid phase and gas phase flow. First liquid propane containing propylene impurity flew from top of the reactor and hydrogen was injected from bottom as a gas phase counter-currently thereafter liquid and gas contacting phases both flew from bottom side of the reactor concurrently. Propylene content of liquid propane was hydrogenated through structured catalyst bed filling the pilot reactor and sampling has been done after establishing steady state conditions. In modeling a set of 12 partial differential equations (PDEs) were developed for dynamic modeling of current process including mass transfer and momentum transfer equations in liquid and gas phase. Afterward orthogonal collocation method was utilized to solve developed PDEs. Finally the results of mathematical modeling showed good consistency with experimental data. Furthermore, the sample analysis showed nil concentration of propylene in countercurrent flow in several operating condition, however nil concentration of propylene was not reported in concurrent flow pattern even by increasing the liquid flowrate.

1. Introduction

Structured packings are well known as the internal devices of column providing superior mass transfer efficiency and low pressure drop (Aroonwilas et al., 2003; Beugre et al., 2011; Owens et al., 2013; Sun et al., 2013). Depending on the physical property of the products structured packings could be fabricated from plastic or metallic materials. Old versions of structured packings were made from metal gauze. The gauze packings which were devised by Sulzer Chemtech first, had huge contribution on distillation technology improvement (Li et al., 2016). Whilst recently metal gauze was changed by metal corrugated which is cheaper (Fair et al., 2000; Sun et al., 2014). Since structured packings offer large effective area ranging 125 (m^2/m^3) to 750 (m^2/m^3), they were broadly accepted in many industrial applications such as dehydration, extraction, distillation and absorption (Aroonwilas et al., 2001; Haroun et al., 2012, 2014; Jokar et al., 2014; Shilkin et al., 2006). Due to the widespread usage of packing beds in columns minor improvement in design could result substantial saving in cost. Therefore many manufacturers attempted to produce more efficient structured packing to diminish the operational and capital costs (Thitakamol et al., 2015).

In recent years many research works focused on evaluation of structured packings performance. Commercial structured packings were attractive enough to be studied in several process conditions. Olujic and Jansen (2015) carried out an experiment with common-size and high-capacity, corrugated-sheet structured packings to find the relation between the quality of liquid distribution and density of initial irrigation profiles. Zakeri et al. (2012) compared the liquid hold up of 3 different types of structured packings: Mellapak 2X from Sulzer, Koch-Glitsch Flexipac 2Y HC and Montz-

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Nomenclature and units

Parameter description

a	Specific surface area (m^2/m^3)
cp	Heat Capacity (J/kmol.K)
C_1-C_5	Dimensionless Constant
C_E	Modification factor (dimensionless)
D	Mass transfer diffusivity (m/s^2)
d_p	Particle diameter (m)
E	Activation Energy (kj/mol)
g	Gravity, 9.81 (m/s^2)
H	Height of the catalyst bed (m)
H_i	Henry's Coefficient of component i (barg)
h	Hold up (dimensionless)
k	Mass Transfer coefficient(m/s)
K	Constant for reaction rate (kmol/hr.kgcat.Pa)
Ke	Constant for equilibrium (Pa^2)
K_2	Dimensionless constant for correlation
MW	Molecular weight (dimensionless)
P_i	Partial pressure of component i (barg)
ΔP	Pressure drop (Pa)
Q	Flow rate (m^3/hr)Q
r	Reaction rate (kmol/kg-catalyst.hr)
R	Gas global constant (kj/kmol.K)
S	Side length of corrugation (m)
T	Temperature (K)
T_c	Critical Temperature (K)
T_r	Reduced Temperature (T/T_C)
t	Time (s)
u	Superficial velocity (m/s)
z	Compressibility factor (dimensionless)

Greek letters

μ	Viscosity (Pa.s)
λ^{eff}	Effective Thermal conductivity (w/m.K)
ρ_m	Molar Density (kmol/m^3)
ρ_b	Catalyst bed bulk density (kg/m^3)
ρ	Mass density (kg/m^3)
ε_b	Void fraction of catalyst bed (dimensionless)
ε	Void fraction (dimensionless)
γ	Shape factor (dimensionless)

Subscripts and Superscripts

b	Bulk
d	Dry
e	Effective
g	Gas Phase
i	Numerator for component
in	Inlet
j	Numerator for component
L	Liquid Phase
p	Particle

Pak B1-250M. Olujic et al. (2012) used new version of J. Montz corrugated sheet structured packings i.e. B1-250MN, B1-350MN and B1-500MN under total reflux distillation to study the effect of dimensions and design of corrugations and the inclination angle on pressure drop. Zhang et al. (2011) employed chromochemical reactive mass transfer technique to study local mass transfer of commercial structured packings with corrugation angle of 45°. Olujic et al. (2015) compared maximum useful capacity of B1-250MN and B1-350MN under total reflux distillation in a pilot column. Ratheesh and Kannan (2004) studied the flexibility of KATAPAK®-SP in variable ratio of the reaction zone relative to the separation zone. Yang et al. (2015) utilized structured packings in the absorption of CO_2 to investigate the effect of packing geometry and flowrate on mass transfer area. Rejl et al. (2015) studied mass transfer characteristics of M250Y, M350Y, M452Y and M500Y under absorption conditions. The recognized privilege of structured packings convinced the researchers to use them in catalytic reactions as the base frame of catalyst. Depend on the characteristics of catalysts some of them could be coated on the foundation of structured catalyst. Italiano et al. (2016) used dip-coating into acid free catalyst dispersion method to synthesize two different type of structured catalyst from Ni/CeO_2 and Ni-Rh/CeO_2 catalyst powder. They washcoated the catalyst powder on the structure of monolith structured packing. Ahmadi et al. (2016) utilized SBA-15, MCM-41, HZSM-5, γ-Al_2O_3 as nano-structured materials to support CoMo catalyst for producing light oil (LO) and heavy oil (HO) with lower O/C and higher H/C.

Natural gas comprises of a variety of hydrocarbons that are used majorly as an energy source like methane and natural gas liquids (NGLs) that have higher value as separate products. Propane is one of the NGLs constituting 3–4% of natural gas that is separated from other NGLs and is used mainly to produce polyolefins. The latest data provided by IHS Chemical North American Propylene Supply Study shows that compared to present situation the market for propylene is projected to grow about 45% by 2023 reaching 130 million metric tonnes per year worldwide. Since the propane acts also as an inert gas in the polymerization reactors producing polyolefins, it should pass through hydrogenation reactor to change any propylene impurity to propane, because propylene impurity could cause side reaction in polymerization reactor that causes temperature run away. Thus propylene hydrogenation is an important industrial process that is used by many licensors through none efficient conventional catalyst bed in many petrochemical plants in several countries. Ahmadigoltapeh et al. (2015) applied fabricated structured and random catalyst of copper-magnesia to study propylene hydrogenation performance through structured and random catalyst bed. Gonzalez-Castano et al. (2016) deposited original powder of gold and platinum onto metallic monolithic structures to compare gold and platinum structured packing in water gas shift (WGS) reaction. Montebelli et al. (2014) employed structured catalyst of washcoated copper honeycomb monoliths to optimized methanol synthesis in multi tubular fixed bed reactor. The effect of Ce and H_2S on selective hydrogenation of butadiene in the presence of benzene was studied by Tailleur and Nascar (2012). They applied NiPdCe(x)/Si-Al coated structured catalyst in a continued stirred tank reactor. Alkylation of benzene with propylene through BeiHua (BH) structured catalytic packing was studied by Dai et al. (2013). They used structured catalyst in a bubble-point reactor and they used CFD modeling to estimate the optimized operating condition. Vervloet et al. (2013) compared structured catalyst with conventional dumped packed bed in a multi-phase tubular fixed reactor for intensification process. They used one-dimensional numerical model to analyze the experimental process results.

This paper concentrated on the aspect that has not been experienced comprehensively so far. The framework of this investigation was studying propylene hydrogenation as an important industrial reaction for the first time under two different flow