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Study of the countercurrent–concurrent gas–liquid flow configuration impact on ethylene hydrogenation within structured catalyst bed: Experiment and modeling

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ABSTRACT

In this paper catalytic hydrogenation of ethylene was studied using liquid propane with 1.5% of ethylene content and hydrogen gas in 30 barg trickle bed reactor under two flow configurations: countercurrent and concurrent. Trickle bed experiments were then performed using an experimental package comprising high pressure pilot reactor, gas chromatograph apparatus, dosing pump and required instrumentation system to compare countercurrent and concurrent sampled data furthermore structured catalyst of alumina–nickel was used as catalytic bed. Trickle bed modeling was achieved using a package of 12 dimensionless two-dimensional homogeneous mass and momentum transfer partial differential equations, the model parameters and kinetic equations. Orthogonal collocation technique was employed to solve developed PDEs. Predicted performance of the trickle bed by mathematical modeling was compared by experimental results and there was good consistency between them. The experimental results showed zero concentration of ethylene through countercurrent flow. Finally, the experimental analysis reports emphasize on zero concentration of ethylene in output of countercurrent bed whilst increasing the Reynolds number did not conclude to nil concentration of ethylene in output of concurrent bed.

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1. Introduction

Recently, environmental issues such as the greenhouse effect and acid rain have started criticisms from the public urging international observatory organizations to plan more restricted regulations on pollution discharge. Since the pollutants are more or less water-soluble, packed beds, because of their cheap operation, are demanding devices for carrying out pollution abatement tasks. The ongoing demand in the pursuit of cost effective units is permanently imposing the necessity for well-designed and highly-efficient packings. This loss will be retrieved by respond to industrial economic requirements (*i.e.* low pressure drop, high mass-transfer efficiency and higher specific area) and simultaneously, to comply with the stringent discharged pollutants. In contrast to the traditional randomly packing materials, structured shape packings identified as an efficient and broadly used contacting devices. Structured shape packings are used in wide range of chemical process such as: distillation, reactive distillation, absorption and dehumidification [1-4]. Their high void fraction provides lower pressure drop and higher capacities, whereas their high surface area provides higher efficiency in separation [5-7]. Recognized superior privileges of structured packings are gaining interest of recent research works to utilize structured shape packings as the basic foundation for catalyst particles [8-10]. In fact typical advantages of these macro porous structures as the structured catalyst pursue from high internal accessibility thanks to high surface area and low pressure drop [11].

1.1. Literature review

One of the most used industrial reaction process using structured catalyst is reactive stripping. Schildhauer et al. compared empirically stacked film-flow monoliths and Sulzer KATAPAK-S in reactive stripping as the catalyst support [12]. In this experiment inert nitrogen met liquid phase counter-currently, stacked monoliths showed higher performance than KATAPAK-S. Afterward in the other independent investigation Schildhauer et al. attempted to model reactive stripping counter-currently in film flow monoliths filled reactor [13]. They also validated the model for a binary and

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Notation

a	specific surface area (m^2/m^3)
C_P	heat capacity (J/kmol·K)
C_{Li}	concentration of component i in liquid phase ($kmol/m^3$)
C_{gj}	concentration of component j in gas phase ($kmol/m^3$)
C_1–C_5	constant (dimensionless)
C_E	modification factor (dimensionless)
D	mass transfer diffusivity of component in gas mixture (m/s^2)
D_{im}	mass transfer diffusivity of component in gas mixture (m/s^2)
d_p	the diameter of particle (m)
E	activation energy (kJ/mol)
g	gravity acceleration (m/s^2)
H	catalyst bed height (m)
H_i	Henry's coefficient of component i (barg)
h	hold up (dimensionless)
k	mass transfer coefficient (m/s)
K	reaction rate constant (kmol/h·kgcat·Pa)
Ke	equilibrium constant (Pa^2)
K_2	correlation constant (dimensionless)
M	molecular weight
P_i	partial pressure of component i (barg)
ΔP	pressure drop (Pa)
Q	flow rate (m^3/h)
r_p	reaction rate (kmol/kg-catalyst·h)
R	gas global constant (kJ/kmol·K)
S	side dimension of corrugation (m)
T	temperature (K)
T_c	critical temperature (K)
T_r	reduced temperature (T/T_C)
t	time (h)
u	superficial velocity of liquid phase through the packed bed (m/s)
z	compressibility factor (dimensionless)

Greek letters

μ	viscosity (Pa·s)
λ^{eff}	effective thermal conductivity (w/mK)
ρ_m	molar density ($kmol/m^3$)
ρ_b	catalyst bed bulk density (kg/m^3)
ρ	mass density (kg/m^3)
τ	tortuosity
ε_p	porosity of one catalyst particle (dimensionless)
ε_b	void fraction of catalyst bed (dimensionless)
ε	void fraction (dimensionless)
γ	shape factor of pellet (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

multi-component system. Shilkin and Kenig proposed an analogy between flow pattern and film flow based on the film flow concept [14]. They verified the proposed model via binary distillation in a column filled with Montz-Pak A3-500. Thereafter Shilkin et al. presented a hydrodynamic approach for temperature and composition profile prediction through reactive stripping in distillation column [15]. In fact they improved the former proposed model for higher gas loads. Finally the model was verified against experimental data on distillation of binary and ternary mixtures. To simulate the gas–liquid separation process Brinkmann et al. introduced a novel modeling method for hydrodynamic analogy (HA) which takes packing geometry directly into account, so experimental estimation of mass transfer coefficient will not be required [16]. They used different types of catalyst coated support internals for esterification of hexanoic process as the test method for model data validation.

Recently some research works focused on making a novel structured type catalyst as a convenient substitution for conventional type. Deeng et al. synthesized a structured catalyst of Cu-ZSM-5 by using hydrothermal method. To make the catalyst, zeolite was coated on stainless steel wire mesh. They studied selective catalytic reduction of NO over synthesized catalyst and reported 94% reduction of NO and 4.6% SCR-HC selectivity [17]. Giani et al. selected the metal foams as the support of structure catalyst. To produce the structured catalyst the metal foams were coated with thin layer of palladium–alumina. They applied the catalyst in a 9-mm diameter tubular reactor by performing the catalytic oxidation of CO in empty tube. Finally they derived a generalized engineering correlation to predict mass transfer coefficient [18]. Guo et al. used sputtering technique to deposit Au, Pt and Au–Pt alloy on porous steel fiber matrices. They reported that thanks to using sputtering technique heterogeneous catalyst was produced clean and fast as reducing agents, solvents or noble metal complexes were not required [19]. Other different techniques of settling catalyst on structured shapes such as suspension, electrophoretic, electrochemical and CVD method were summarized in a review paper by Meille [20]. Tailleur et al. studied the effect of H_2S and Ce on selective hydrogenation of diolefins. They applied NiPdCe(x)/Si–Al-coated structured packing catalyst for investigation of butadiene hydrogenation in the presence of benzene. They proved that the electronic configuration and metal dispersion both were affected by H_2S and Ce [21]. This fact presented the modified selectivity and activity of the structured catalyst at constant Ni and Pd contents.

Some researchers compared the novel structured type catalyst beds with conventional one through experiment and mathematical simulation. Ivanova et al. deposited ZSM-5 coatings on β-SiC monolith foams to synthesize the structured catalyst. Then they compared structured catalyst bed with conventional one within an experiment. In comparison with conventional bed they found considerable activity/selectivity reaction improvements in conversion of methanol to olefin through structured catalyst bed [22]. Wang et al. selected VOC catalytic combustion as a target reaction to compare a new designed platinum-structured catalyst on anodic aluminum support with traditional catalyst. The results of experiment and modeling both concluded that structured catalyst improved mass transfer efficiency and showed higher catalytic activity [23]. Ahmadigoltapeh et al. compared new designed structured catalyst of copper–magnesia with conventional one through hydrogenation reaction. They found higher mass transfer efficiency and more compatibility with high liquid phase velocity in structured catalyst bed [24]. Maestri et al. opted reaction of CH_4 partial oxidation to compared different structured catalyst supports namely foams, honeycomb monoliths, square channels and spheres via experiment and modeling. They realized that the mass and heat transfer properties have serious effect on reaction at transient and steady state condition [25].

Many research and studies have been conducted in the field of structured catalyst, however most of them carried out with academic fluids or at ideal conditions like low pressure. Since the findings of the research works on structured packing will be used

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