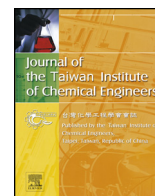




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Dimethyl ether synthesis in a gas–solid–solid trickle flow reactor with continuous adsorbent regeneration

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ABSTRACT

Dimethyl ether (DME) as a green fuel is commercially produced by methanol dehydration in an adiabatic fixed bed reactor. This work describes a mathematical modeling and simulation of DME reactor in which Zeolite 4A, with the composition of $\text{Na}_{12}(\text{Si}_{12}\text{Al}_{12}\text{O}_{48})\cdot 27\text{H}_2\text{O}$, is considered as water adsorbent. For this purpose, a gas–solid–solid trickle flow reactor (GSSTFR) is used instead of conventional DME synthesis reactor (CR). The main advantage of GSSTFR over the conventional sorption-enhanced reaction process is the continuous adsorbent regeneration in this novel configuration. The result demonstrates that selective adsorption of water from DME synthesis in the proposed configuration leads to 11.17% enhancement in DME production rate compared to CR. Subsequently, the effect of operating conditions on the performance of the aforementioned reactor is investigated. This paper shows how the concept of in-situ water adsorption is feasible and beneficial for dimethyl ether synthesis.

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1. Introduction

Natural gas as the cleanest and most abundant fossil fuels is currently one of the most promising topics in energy industry. With the growing global markets for natural gas, it is important to identify effective methods to deploy the vital resource worldwide. Dimethyl ether (DME) which is produced by the conversion of various feedstock such as natural gas, coal, oil residues and biomass into syngas has attracted wide attention as an ultraclean fuel, because it produces no SO_x and minimal NO_x and CO emissions [1]. It is also an easy substitute for liquefied petroleum gas (LPG), owing to its similar properties to LPG [2]. DME is a useful chemical intermediate for the preparation of many important chemicals, such as methyl acetate and lower olefins [3].

Usually, high purity DME is synthesized by dehydration of methanol produced from syngas, involving two-step processes: methanol synthesis over a copper-based catalyst and then methanol dehydration over a γ -alumina catalyst or zeolites in order to produce DME. Also recently the direct conversion of syngas over hybrid catalysts containing two functionalities, *i.e.*

metallic for methanol production and acid for methanol dehydration have been developed for DME synthesis [1,2,4]. Mathematical modeling of catalytic DME synthesis reactors has been the subject of numerous studies [5–11]. A complete literature review on DME synthesis has been done by Azizi et al. [12].

Recently, the concept of contacting gas and fine solid particles (flowing solids) inside a packed bed, has been proposed to solve reactor problems. In this type of equipment, fine solid particles (adsorbents) and gas flow counter-currently (or co-currently) through the column packed with other solid phase (catalysts). The fluid dynamics of such systems received considerable attention over the years, and this included mass and heat transfer studies. In the literature, such columns are under the name of gas-flowing solids-fixed bed contactors, or, gas–solid–solid trickle flow contactors, or raining packed bed contactors.

The concept of contacting gas and fine solids particles (flowing solids) inside a fixed bed was patented nearly sixty years ago [13]. The interest in exploiting the unique features of the contacting gas and fine solid particles was enhanced by the studies of Westerterp and colleagues [14]. Kuczynski et al. [15] have investigated an experimental study on methanol synthesis in a counter-current gas–solid–solid trickle flow reactor. The results showed that the conversion of reactant was higher when the product was removed from the reactor. In 1988, the intensified process was compared to the conventional methanol synthesis in an optimized Lurgi process

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Nomenclature

A_c	cross section area of each tube (m^2)
Ar	Archimedes number for flowing solid particles, ($= d_p^3 \rho_g (\rho_p - \rho_g) g / \mu^2$)
a_s	specific surface area of catalyst pellet (m^2/m^3)
a'_s	specific surface area of flowing adsorbent (m^2/m^3)
C_d	drag coefficient
$C p_g$	specific heat of the gas at constant pressure (J/(mol K))
$C p_s$	specific heat of the catalyst at constant pressure (J/(mol K))
$C p'_s$	specific heat of the flowing adsorbent at constant pressure (J/(mol K))
c_t	total concentration (mol/ m^3)
D_i	tube inside diameter (m)
D_{ij}	binary diffusion coefficient of component i in j (m^2/s)
D_{im}	diffusion coefficient of component i in the mixture (m^2/s)
d_{eq}	equivalent diameter of packing particles ($= 6(1 - \varepsilon)/(a_s + 4/D)$) (m)
d_s	catalyst diameter (m)
d'_s	flowing adsorbent diameter (m)
F_i	molar flow of species i (mole/s)
F_t	total molar flow per tube (mole/s)
G	mass flux of gas ($kg/(m^2 s)$)
ΔH_{ads}	specific heat of adsorption (J/mol)
$\Delta H_{f,i}$	enthalpy of formation of component i (J/mol)
ΔH_{298}	enthalpy of reaction at 298 K (J/mol)
h_f	gas-catalyst heat transfer coefficient (W/($m^2 K$))
h'_f	gas-adsorbent heat transfer coefficient (W/($m^2 K$))
k_1	rate constant of dehydrogenation reaction ($mol^2/(m^3 kg s)$)
K	conductivity of fluid phase S (W/m K)
K_{eq}	reaction equilibrium constant for methanol dehydration reaction (mol/m^3)
K_i	adsorption equilibrium constant for component i (m^3/mol)
k_{gi}	gas-catalyst mass transfer coefficient for component i (m/s)
k'_g	gas-adsorbent mass transfer coefficient (m/s)
L	length of reactor (m)
M_i	molecular weight of component i (g/mol)
N	number of components used in the model ($N = 3$)
P	total pressure (bar)
q	concentration of water adsorbed in flowing adsorbent (mol/kg)
q_e	equilibrium concentration of adsorbed water (mol/kg)
R	universal gas constant (J/(mol K))
Re_s	Reynolds number of packing elements (–)
Re'_s	Reynolds number of flowing adsorbent (–)
r_i	reaction rate of component i (mol/(kg s))
S	mass flux of flowing adsorbent ($kg/(m^2 s)$)
T_g	bulk gas phase temperature (K)
T_s	temperature of catalyst phase (K)
T'_s	temperature of flowing adsorbent (K)
U_g	real gas velocity, ($= G/\rho_g \varepsilon'$) (m/s)

u_g	superficial gas velocity (m/s)
u_r	relative velocity for co-current flow of gas and flowing adsorbent, ($= U_g - u'_s$) (m/s)
u'_s	real flowing adsorbent velocity, ($= S/\rho'_s \beta$) (m/s)
y_i	mole fraction of component i in the fluid phase (mol/mol)
y_{is}	mole fraction of component i in the catalyst phase (mol/mol)
z	axial reactor coordinate (m)

Greek letter

β	flowing solids holdup, ($= \beta_d + \beta_s$)
β_d	dynamic flowing solids holdup
β_s	static flowing solids holdup
ε	void fraction of catalytic bed (m^3/m^3)
ε'	void fraction corrected due to presence of the flowing adsorbent, ($= \varepsilon - \beta$) (m^3/m^3)
ϕ	sphericity of packed bed element
η	effectiveness factor
μ	dynamic viscosity (Pa s)
ρ_B	catalytic bed density (kg/m^3)
ρ_g	gas density (kg/m^3)
ρ_s	catalyst density (kg/m^3)
ρ'_s	flowing adsorbent density (kg/m^3)

[16]. Recently, Bayat and colleagues have investigated the use of gas-flowing solids-fixed bed reactors for methanol synthesis [17] and gasoline production [18] with the purpose of maximizing production rates and selectivity.

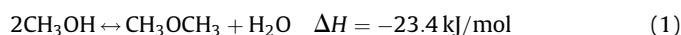
Zeolite 4A which is applied as the adsorbent is a white solid particle, with the composition of $Na_{12}(Si_{12}Al_{12}O_{48}) \cdot 27H_2O$. It has high capacity for water adsorption which makes it interesting in water removal or separation [19]. The regeneration of zeolite 4A is based on desorption of water vapor. The dehydration of zeolites is usually conducted under vacuum, or in a flow of the carrier gas, with a simultaneous raising of the temperature to 573–673 K [20]. Zeolite 4A thermal stability is rather good and only above 1073 K the change of its structure occurs which is accompanied with drop in water capacity [21].

2. Objectives

In this work, the considered Process Intensification idea would be to introduce a third solid phase as regenerative flowing adsorbents to the conventional reactor. The underlying goals of this study are to develop a steady-state, one dimensional model for dimethyl ether synthesis in a gas–solid–solid trickle flow reactor (GSSTFR) with in-situ water removal to improve DME production and decrease catalyst deactivation. The operating condition of an actual adiabatic packed bed reactor has been considered as a basis for simulating the GSSTFR. Afterward, the simulation results are compared with the ones in the conventional reactor. To the best of our knowledge, the GSSTFR with in-situ water adsorption has not yet been applied for methanol dehydration process.

3. Reaction scheme and kinetics

Conventionally, DME is synthesized by dehydration of methanol. The reaction is exothermic and expressed as follows [21–25]:



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