



Investigation and recovery of purge gas streams to enhance synthesis gas production in a mega methanol complex



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ABSTRACT

Nowadays, methanol has gained great importance as an alternative fuel and a product which can be considered as a CO₂ utilization in novel processes; nevertheless, there is no adequate work on purge gas streams in current conventional methanol complexes. This study aims to specify main waste gasses in one of the biggest methanol plants in the world, located in the Pars Special Economic Energy Zone of Iran. The calculations in this work show that every hour, 50106 Sm³ purge gas including unreacted gas (60% hydrogen, 17% CO and CO₂, and 23% N₂ and CH₄) is burnt in the complex. A new process design has been proposed to recovery of available purge gasses in that methanol plant. The Auto-Thermal Reformer (ATR) of the complex has been modeled in detail and various cases were considered to analyze the effects of this new strategy. The results indicate noteworthy improvements in both quality and quantity of the produced synthesis gas by injecting the recoverable stream into the ATR. In this way, emission of 26.7 ton carbon dioxide per hour to the atmosphere is prevented and it converted to valuable methanol. This process can bring annual profit ranging from 20 to 40 million US dollars per year with the Rate of Return (ROR) between 29 and 56%.

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1. Introduction

CO₂ is considered as one of the main greenhouse gasses and proved to be the primary cause of global warming issue. Among various sources for CO₂ emission, flare and purge gas are known as one of the major ones. The IPCC (Intergovernmental Panel on Climate Change) has reported that in the beginning of twenty century, CO₂ emission from fossil fuel, cement, and flaring was 4 Gt/year which rose to 27 and 37 Gt in 2000 and 2014, respectively [1]. Purging and flaring gas are an inevitable part of almost all petrochemical complexes: In many purification cases, undesirable dissolved gasses in the liquid phase are removed by depressurizing and sent to the flares. In other situations, purge gas streams are used commonly to control processes under steady state operation. For example, in ammonia synthesis loop, a percentage of the process gas is continuously purged to preserve concentration of the inert gas (argon, methane, etc.) under the defined level [2].

To date, various approaches have been suggested in order to prevent CO₂ from entering the atmosphere. One of the novel proved approaches to mitigate the ever-growing rate of climate

change is Carbon Capture and Storage (CCS) technologies [3]. In another promising approach, CO₂ becomes the feedstock to produce useful products rather than a waste which needs to be sequestered. As CO₂ has a low-grade energy from thermodynamic perspective, its conversion to hydrocarbons needs consumption of energy. A new method is using solar energy to infuse CO₂ molecules [4]. Catalysts used in such a process are named photocatalyst that most famous one is titanium dioxide because of its low cost, high thermal and chemical stability [5,6]. Although photocatalytic reduction of CO₂ is very attractive due to the ultimate availability of solar energy, the practical photocatalysts with enough efficiency are bottlenecks [7]. Another method is CO₂ hydrogenation in which the reduction occurs mainly in high temperature and presence of water. This method can be applied to produce various hydrocarbons such as formic acid, carbon mono oxide, methane and especially methanol [8,9].

Even though in recent years, methanol production has gained great importance due to its potential role in mitigating and using anthropogenic CO₂, conventional methanol plants are still using purge gas streams both in purification and synthesis processes and there are no sufficient studies on these purge streams and their values. The purpose of this study is to investigate flare and purge gasses in one of the world's biggest methanol complexes located in the south of Iran. According to National Geophysical Data Center

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Nomenclature

A_c	Cross section area of each tube (m ²)
A_o	$k - \varepsilon$ model constant
A_s	$k - \varepsilon$ model constant
C_p	Specific heat of bulk gas at constant pressure (J mol ⁻¹ K ⁻¹)
$C_{p,i}$	Specific heat of the species i at constant pressure (J mol ⁻¹ K ⁻¹)
$C_{1\varepsilon}$	$k - \varepsilon$ model constant
C_2	$k - \varepsilon$ model constant
C_μ	$k - \varepsilon$ parameter
d_p	Particle diameter, m
E_j	Activation energy of reaction j , J mol ⁻¹ ($j = 10-12$)
F_i	Mole flow rate of species i (mol s ⁻¹)
f	Mixture fraction
G_k	Generation of turbulent kinetic energy due to mean velocity gradients
G_b	Generation of turbulent kinetic energy due to buoyance
H	Standard enthalpy, J mol ⁻¹
$\Delta H_{R,j}$	Heat of reaction j ($j = 10-12$), J mol ⁻¹
ΔH_{298°	Standard heat of reaction, J mol ⁻¹
k	Turbulent kinetic energy
K_{ej}	Equilibrium constant of reaction j
$K_{o,i}$	Constant
$K_{o,j}$	Constant
p	Total pressure (Pa)
p_i	Partial pressure of component i (bar)
Q_R	Radiation heat loss
R_j	Rate of reaction j kmol/kgcat. h
r_i	Conversion rate of species i , kmol/kgcat. h
S	Modulus of the mean rate-of-strain tensor
T	Temperature (K)
U_g	Linear velocity of gas phase (m s ⁻¹)
w_i	Reaction rate of species i
Y_i	Mass fraction of species i
z	Axial reactor coordinate (m)

Greek letters

ξ	Void fraction of catalytic bed
ε	Rate of dissipation of the turbulent kinetic energy
η	Effectiveness factor
η'	$k - \varepsilon$ model parameter
ρ	Bulk gas density, kg/m ³
ρ_b	Catalyst bed reactor, kg m ⁻³
μ	Dynamic viscosity of gas, Pa.s
ν	Kinematic viscosity, m ² /s
χ	Scalar dissipation rate
σ_k	$k - \varepsilon$ model constant
σ_ε	$k - \varepsilon$ model constant

Subscript

g	Gas
I	X- direction
i	Component numerator
J	Y- direction
j	Reaction numerator
<i>Out</i>	Outlet

Superscript

C	Combustion chamber
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Abbreviations

ATR	Auto-Thermal reformer
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CCS	Carbon capture and storage
PSA	Pressure swing adsorption
SN	Stoichiometric number

(NGDC), Iran ranks third in applying gas flaring among all countries [10]. Petrochemical industries are the third highest energy intensive industries in Iran, having a share of 16% of the total energy consumption [11]. As a result, efficiency enhancements in this sector would significantly help reduce giving off pollution by that country. This work detects potential recoverable streams in that plant and investigates injection of these streams to the Auto-thermal Reformer (ATR) as a proposed method to increase efficiency and methanol production in an environmentally friendly way. To comprehensive effects of this recovery on performance of the ATR, this reactor has been modeled and different cases are discussed.

2. Process description and purge gas streams recovery

This methanol complex consists of twine methanol plants which each has a capacity of producing 5000 ton methanol with grade AA per day. The schematic diagram of the production process is shown in Fig. 1 and the three main units are explained briefly to study purge gas streams and overall process in following subsections.

2.1. Reforming unit – unit 100

The reforming unit aim is converting of natural gas feedstock to synthesis gas. This unit is a combined arrangement of a steam reformer and an auto-thermal reformer. In the beginning of this unit, desulfurized and saturated natural gas divided into two streams. One stream is used as the ATR feed, and another one passes first through a pre-reformer, and then through the tubular reformer. In the tubular reformer, tubes are located in a rectangular furnace with the firing system on its top to provide the required heat for high endothermic reactions. The heat duty estimation for the process can be found in [12]. The outlet of the tubular reformer forms another feed of the auto-thermal reactor. The schematic diagram of the reforming unit is displayed in Fig. 2.

2.1.1. Auto-thermal reformer

Generally, an Auto-thermal Reformer is composed of two main sections: homogenous section (combustion chamber) and heterogeneous section (catalytic bed) (Fig. 3). The first section consists of a burner and a thermal zone. The heat duty of the reactor is supplied in a burner by combustion reactions. Beneath the burner a thermal zone is located where reforming reactions take place homogeneously, causing the gas temperature to decrease.

ATRs feeds are hydrocarbons (process syngas stream), steam and oxidizer. Oxygen, as an oxidizer, is an important feed of auto-thermal reactors which is contributed to 40% of their operating cost [13]. In an ATR feed, oxygen to carbon (O/C) ratio affects the outlet temperature, composition and coke formation along the reactor. This ratio is usually considered slightly more than the optimum value for any possible heat loss [14]. Increasing the amount of oxygen promotes methane conversion and the yield of hydrogen, while its excess reduces hydrogen production, as in this way, CH₄ tends to be converted to H₂O and CO₂ rather than H₂ and CO [15,16]. Another feed of auto-thermal reformers is steam as a reactant in both reforming and water gas shift reactions. In literature, it has been shown that H₂O/CH₄ > 1 in association with a temperature greater than 900 °C can eliminate the soot formation in ATRs [17].

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