



A polygeneration from a dual-gas partial catalytic oxidation coupling with an oxygen-permeable membrane reactor



Yanhong Hao^{a,b,c}, Yi Huang^{a,b}, Minhui Gong^{a,b}, Wenying Li^{a,b,*}, Jie Feng^{a,b}, Qun Yi^{a,b,*}

^aKey Laboratory of Coal Science and Technology (Taiyuan University of Technology), Ministry of Education and Shanxi Province, Taiyuan 030024, PR China

^bTraining Base of State Key Laboratory of Coal Science and Technology Jointly Constructed by Shanxi Province and Ministry of Science and Technology, Taiyuan 030024, PR China

^cEnvironmental Engineering Department, Shanxi University, Taiyuan 030013, PR China

ARTICLE INFO

Article history:

Received 24 July 2015

Accepted 22 September 2015

Keywords:

Polygeneration

Coke oven gas

Coal gasified gas

Partial oxidation

Oxygen-permeable membrane

ABSTRACT

Polygeneration system, typically involving chemicals/fuels and electricity co-production, is a promising technology for the sustainable development of energy and environment. In this study, a new polygeneration system based on coal and coke oven gas (COG) inputs for co-production of dimethyl ether (DME)/methanol and electricity is proposed. In the new system, an appropriate syngas for the synthesis of DME is from coal gasified gas (CGG) reforming of COG coupled with an oxygen-permeable membrane reactor, in which both COG and CGG reforming process and fuel combustion process are incorporated, which reduces exergy destruction in the whole reforming process. In order to obtain the best performance of CO₂ reduction, energy saving and economic benefit, the key operation parameters of the proposed process are analyzed and optimized. The new system is compared with the process based on CH₄/CO₂ dry reforming, in terms of exergy efficiency, exergy cost and CO₂ emissions. Through the new system, the exergy efficiency can be increased by 7.8%, the exergy cost can be reduced by 0.88 USD/GJ and the CO₂ emission can be reduced by 0.023 kg/MJ. These results suggest that the polygeneration system from CGG and COG partial catalytic oxidation coupling with an oxygen-permeable membrane reactor (PL-PCO-OPMR) would be a more attractive way for highly efficient and clean use of CGG and COG.

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1. Introduction

As one of the most promising technologies for clean and efficient use of energy today, polygeneration system synergistically integrating the chemical engineering and power generation systems can realize cascade utilization of both chemical and thermal energies, and step-by-step conversion of effective components [1]. In addition, polygeneration system is a highly flexible and cross-sector integrated system of resources, energy and environment, which can simultaneously make two or more marketable products, such as chemicals, fuels and electricity from a mix of different energy resources with element and energy efficiencies and pollution emission reduction, all of which are potentially higher than current standalone practices [2–4]. Base on these advantages, a number of polygeneration systems on the basis of different energy resources types (such as coal, natural gas, solar and biomass) with

various configurations and products (eg. methanol (MeOH), dimethyl ether (DME), dimethyl carbonate (DMC), Fischer–Tropsch oil and electricity) are proposed and investigated [5–10].

As the biggest coke production country, nowadays China produces about 500 million tons of coke, with the resulting by-product of about 210 billion m³ of coke oven gas (COG, 6 vol% CO, 59 vol% H₂, 26 vol% CH₄, 3 vol% CO₂, and 6 vol% N₂) which is a presently underutilized hydrogen-rich product [11]. Except for about 50% of that is COG recycled to provide energy for coking process, most of the rest is discharged into the air after combustion, leading to a waste of potential energy resources and also causing serious air pollution. Hence, it is necessary to find a suitable way for high efficient and clean utilization of COG resources. Since COG is rich in hydrogen and methane, we can use COG to reform with carbon-rich gas such as CO₂, CGG (coal gasified gas) and flue gas etc. via CH₄/CO₂ dry reforming to produce syngas at a proper ratio of C/H for downstream production [12–14], and syngas is an indispensable medium for the synthesis of chemicals and liquid fuels in energy chemical engineering.

Bermúdez et al. [14,15] investigated CO₂ reforming of COG over Ni-based catalysts to produce syngas for methanol synthesis, and

* Corresponding authors at: Key Laboratory of Coal Science and Technology (Taiyuan University of Technology), Ministry of Education and Shanxi Province, Taiyuan 030024, PR China. Tel.: +86 351 6018957; fax: +86 351 6018453.

E-mail address: ying@tyut.edu.cn (W. Li).

Nomenclature

Capital letters

BCFNO	$\text{BaCo}_{0.7}\text{Fe}_{0.2}\text{Nb}_{0.1}\text{O}_{3-\delta}$
C_{DME}	concentration of DME, mol/m ³
$C_{\text{H}_2\text{O}}$	concentration of H ₂ O, mol/m ³
C_{MeOH}	concentration of methanol, mol/m ³
C_Q	the cost of heat transfer, USD/GJ
C_w	the cost of power input, USD/GJ
$E_{k_{in}}$	the exergy of input of the stream k , MW
$E_{k_{out}}$	the exergy of output of the stream k , MW
EX_{fuel}^{in}	the total exergy of input fuel, MW
EX_{ch}^{out}	exergy of the chemical product, MW
EX_p^{out}	exergy of the output work, MW
$F_{\text{CH}_3\text{OH}}$	methanol outflow rate of products, kmol/h
F_{DME}	dimethyl ether outflow rate of products, kmol/h
$(F_{\text{CO}} + F_{\text{CO}_2} + F_{\text{CH}_4})_{in}$	the total CO, CO ₂ and CH ₄ inflow rate of system, kmol/h
$(F_{\text{CH}_4} + F_{\text{CO}_2})_{out}$	CO ₂ and CH ₄ outflow rate of synthesis unit, kmol/h
I_k	the investment of the unit k , USD
K_{CO}	adsorption constant of CO, bar ⁻¹
K_{CO_2}	adsorption constant of CO ₂ , bar ⁻¹
K_{H_2}	adsorption constant of H ₂ , bar ⁻¹
$K_{\text{H}_2\text{O}}$	adsorption constant of H ₂ O, bar ⁻¹
K_{MeOH}	adsorption constant of methanol, bar ⁻¹
$K_3^0, K_4^0, K_5^0, K_6^0$	thermodynamic equilibrium constant of the reaction (1–4)
N	plant life, year
PL-DR	polygeneration system from CGG and COG based on CH ₄ /CO ₂ dry reforming
PL-PCO-OPMR	polygeneration system from CGG and COG partial catalytic oxidation coupling with an oxygen-permeable membrane reactor
PV_f	remanent value of equipment
S_r	purchase cost of facility in the reference scale, USD
X	the scale of proposed plant
X_r	scale of facility in the reference scale,
U_C	carbon element utilization of system
$U_{\text{CH}_4+\text{CO}_2}$	CO ₂ + CH ₄ conversion efficiency of system

Lowercase letters

$c_{k_{in}}$	exergy cost per unit of input fuel, USD/GJ
f_{CO}	fugacity of CO in gas phase, bar
f_{CO_2}	fugacity of CO ₂ in gas phase, bar
f_{H_2}	fugacity of H ₂ in gas phase, bar
$f_{\text{H}_2\text{O}}$	fugacity of H ₂ O in gas phase, bar
f_{MeOH}	fugacity of methanol in gas phase, bar
i	the discount rate, %

k_1, k_2, k_3, k_4	reaction rate constant of the reaction (1–4), mol/(s kg bar)
r_1, r_2, r_3, r_4	reaction rate of the reaction (1–4), mol/(s kg)
t	tonne
y	year

Greek letters

α	scale factor of facility in the present scale
ω	the circulation ratio of unreacted gas
λ	the ratio of coke oven gas to coal gasified gas
η	the exergy efficiency of system
μ	the cost coefficient of operation & maintenance
τ	operation time

Acronyms

AAFBG	ash-agglomerated fluidized bed gasifier
ASU	air separation unit
BWRS	method Benedict–Webb–Rubin–Starling equation of state
CGG	coal gasified gas
Compr model	compressor/turbine
USD	United States dollar
COG	coke oven gas
COS	carbonyl sulfur
DMC	dimethyl carbonate
DME	dimethyl ether
Flash model	flash vessel
GTL	gas-to-liquid
Heater model	heater/cooler
Heatx model	two-stream heat exchanger
HRSG	heat recovery steam generator
IGCC	integrated gasification combined cycle
JOGMEC	Japan Oil, Gas and Metals Corporation
LHV	low heat value
M	million
Mcompr model	multistage compressor/turbine
MeOH	methanol
MIEC	mixed ionic and electronic conductor
O&M	operation & maintenance
PR-BM	method Peng–Robinson equation of state with Boston–Mathias modifications
Radfrac columns	rigorous distillation tower
RAU	reforming auxiliary unit
Rcstr	continuous-stirred tank reactor
RStoic	stoichiometric reactor
Sep model	multi-outlet component separator

proposed a new process for producing methanol from COG by means of CO₂ reforming. Compared with a conventional process of methanol synthesis from natural gas, the new process has been proved to be superior to the conventional process from the perspective of environment, raw material exploitation and purification costs, and also consumes less energy than conventional production. Yi et al. [16,17] designed and estimated a polygeneration system for producing MeOH/DME/electricity from COG reforming with CGG, and it is found that the polygeneration system shows better performance in terms of energy efficiency, economy and CO₂ emission compared with the stand-alone production. Similarly, Lin et al. [18] proposed a new polygeneration system for methanol and power production based on COG reforming of

CGG with CO₂ recovery, and the new system presents good thermal performance and CO₂ is also avoided with less energy penalty. Qian et al. [19,20] demonstrated a conceptual design of COG assisted coal to olefins process, and the two co-feed systems ensure great reduction of CO₂ emissions and significantly improve energy efficiency compared with coal-to-olefins process. Generally, Ni-based catalysts are widely used for CH₄/COG reforming due to its properties of low cost and high activity. However, carbon deposition on nickel catalysts is a fatal problem for CH₄/CO₂ dry reforming and is the main reason for not industrialization of this technology [21,22].

In order to solve the carbon deposition problem in CH₄/CO₂ dry reforming, another approach, CH₄/CO₂/O₂ partial catalytic

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