

## Postprint: Study of a Natural Gas Polygeneration System Based on Complementary Biomass-Coal Chemical Looping Combustion

**Authors:** Fan Junming, Hong Hui, Jin Hongguang

**Date:** 2017-10-17T00:00:00+00:00

### Abstract

In response to the technical bottlenecks of inefficient energy utilization and excessively high energy consumption for carbon capture in traditional coal-to-natural gas processes, this paper explores a biomass-coal complementary natural gas power cogeneration system based on chemical looping combustion capable of achieving zero CO<sub>2</sub> emissions. The complementation of biomass and coal adjusts the H<sub>2</sub>/CO ratio in syngas from the gasification source, facilitating the methanation reaction process, while chemical looping combustion enables CO<sub>2</sub> capture with zero energy consumption. The results indicate that the total energy efficiency of the system ( $\eta_{(en)}$ ) is 57.03%, the efficiency ( $\eta_{(ex)}$ ) is 54.65%, the system energy saving rate reaches 18.6%, and zero CO<sub>2</sub> emissions from the system are achieved. The influence of key parameters such as oxygen-to-carbon ratio (O/C), steam-to-carbon ratio (S/C), biomass-coal complementary ratio, and unreacted gas recycle ratio on the thermodynamic performance of the system is analyzed. The viscosity of isooctane and alcohols (including propanol, pentanol, hexanol, heptanol) was calculated, and the calculation results show that the overall average absolute deviation between calculated and experimental values is 1.00%, with a maximum deviation of 9.75%.

### Full Text

#### Preamble

#### Study on a Chemical Looping Combustion-Based Hybrid Biomass-Coal System for Synthetic Natural Gas and Power Co-Generation

FAN Jun-ming<sup>1,2</sup>, HONG Hui<sup>1,\*</sup>, JIN Hong-guang<sup>1</sup>

<sup>1</sup> Institute of Engineering Thermophysics, Chinese Academy of Sciences, Beijing 100190, China

<sup>2</sup> University of Chinese Academy of Sciences, Beijing 100049, China

---

## Abstract

To address the technical bottlenecks of conventional coal-to-synthetic natural gas (SNG) processes—namely, inefficient energy utilization and excessive energy penalties for CO<sub>2</sub> capture—this study investigates a novel coal and biomass co-feed polygeneration system for SNG and power production integrated with chemical looping combustion (CLC) to achieve zero CO<sub>2</sub> emissions. The hybrid biomass-coal feedstock inherently adjusts the H<sub>2</sub>/CO ratio in syngas at the gasification stage, which benefits the subsequent methanation process. The CLC unit enables CO<sub>2</sub> capture with zero energy penalty. Thermodynamic performance analysis reveals that the system can achieve an energy efficiency (η) of 57.03% and an exergy efficiency (η<sub>ex</sub>) of 54.65%, with a primary energy saving rate of 18.6% when 40 wt.% of coal is replaced by biomass. Net CO<sub>2</sub> emissions are expected to be 0 kg/h due to the inherent CO<sub>2</sub> separation capability of CLC. The study further analyzes the influence of key parameters—including oxygen-to-carbon ratio (O/C), steam-to-carbon ratio (S/C), biomass share, and unreacted gas recycling ratio—on system thermodynamic performance.

**Keywords:** coal; biomass; SNG; power

---

## 0 Introduction

China's energy resource endowment is characterized by abundant coal, scarce oil, and limited natural gas, which has historically made coal the dominant component of national energy consumption. Traditional direct coal combustion not only results in low utilization efficiency but also causes severe environmental problems. Meanwhile, China's natural gas supply gap continues to expand annually, with increasing dependence on imports. According to forecasts, by 2020, China's natural gas shortage will reach [MATH\_ERROR] m<sup>3</sup>, and the import dependency ratio will exceed 24% [1]. Given this energy structure, developing coal-to-SNG technology holds significant promise. China's first industrial coal-to-SNG project was completed in 2013, and by the end of that year, four such projects had been approved [2]. However, conventional coal-to-SNG processes suffer from three major drawbacks: (1) The primary objective is maximizing SNG yield, necessitating water-gas shift reactions to adjust the H<sub>2</sub>/CO ratio in syngas, which consumes substantial water resources and causes significant irreversible losses. Additionally, recycling large quantities of unreacted gas to achieve ultra-high methane conversion dramatically increases compression power requirements and irreversible losses in the SNG synthesis unit [3]. (2) Unreasonable heat recovery methods result in insufficient steam-generated electricity to meet the process power demands, requiring external power purchases or additional generation units and reducing overall energy efficiency. (3) Despite

CO concentrations exceeding 30% in coal-to-SNG processes, CO capture still requires substantial energy consumption.

To overcome these limitations, this paper proposes a hybrid biomass-coal poly-generation system integrated with chemical looping combustion. This system explores efficient coal conversion pathways for SNG production while seeking low-cost, low-energy approaches for CO capture, providing potential solutions to the bottlenecks in coal-to-SNG technology development.

---

## 1.1 Process Description

[Figure 1: see original paper] illustrates the proposed SNG and power poly-generation system with chemical looping combustion driven by biomass and coal. Coal and biomass first undergo gasification reactions with oxygen and steam to produce syngas rich in CO and H<sub>2</sub>. The generated syngas passes through heat recovery and gas conditioning before entering a condenser for cooling, followed by a desulfurization unit to remove H<sub>2</sub>S. The cleaned syngas then enters the methanation synthesis unit. The heat released during methanation is recovered via high-temperature superheated steam to drive a steam turbine for power generation. In the methanation unit, the unreacted gas recycle ratio is controlled at a moderate level rather than pursuing complete conversion of CO and H<sub>2</sub>. The produced CH<sub>4</sub>, along with unreacted CO, H<sub>2</sub>, and CO<sub>2</sub>, are separated in the SNG purification unit to generate pipeline-quality synthetic natural gas. Meanwhile, the unreacted gases (CO, H<sub>2</sub>, CO<sub>2</sub>) and trace CH<sub>4</sub> are fed into the fuel reactor of the chemical looping combustion system, where they are completely converted to CO and H<sub>2</sub>O by the oxidized oxygen carrier (NiO). The reduced oxygen carrier (Ni) is regenerated in the air reactor through oxidation with fresh air. The high-temperature flue gases from both reactors sequentially pass through a gas turbine and heat recovery steam generator for power generation and heat recovery. The recovered heat produces superheated steam to drive the steam turbine. Since the fuel reactor exhaust consists solely of CO and H<sub>2</sub>O, water can be easily removed by condensation. The CO is then compressed through three-stage intercooling, dehydrated to remove saturated water, pressurized to 80 bar via a CO pump, and finally transported through pipelines.

---

## 1.2 Main Features

The process flow shown in [Figure 1: see original paper] exhibits several distinctive features:

- 1) Leveraging the high carbon content in coal and high hydrogen content in biomass, the hybrid feedstock adjusts the H<sub>2</sub>/CO ratio in syngas at the gasification stage, reducing exergy losses in the subsequent methanation process. Additionally, substituting biomass for 部分 coal reduces coal

consumption.

- 2) The water-gas shift process is eliminated due to its high water consumption and exergy losses. The H<sub>2</sub>/CO ratio is instead regulated through the biomass-coal blending ratio, reducing water consumption in coal-to-SNG projects.
- 3) Moderate recycling of unreacted gas avoids the sharp increase in compression power and methanation exergy losses associated with excessively high recycle ratios in conventional processes.
- 4) Chemical looping combustion enables zero-energy-penalty CO<sub>2</sub> capture. Unreacted gases enter the CLC unit for inherent CO<sub>2</sub> separation, with the exhaust gases utilized in the combined cycle for power generation.

## 2.1 Simulation Method

The composition characteristics of coal and biomass are presented in Table 1 [4, 5], and the main system parameters are listed in Table 2. This study employs Aspen Plus software for system simulation, using the Peng-Robinson (PR) equation of state. The RYield and RGibbs models in Aspen Plus are adopted to simulate solid fuel pyrolysis and gasification. Based on elemental mass conservation, coal pyrolysis decomposes coal into H, N, S, C, O, and ash components. Given the complex mechanisms of pyrolysis and gasification, an RGibbs equilibrium reactor (based on Gibbs free energy minimization) is used to generate syngas [6, 7]. The Compr model simulates compression and expansion processes, while component splitters (Sep) model the air separation, desulfurization, and SNG purification units, with energy consumption calculated based on actual process requirements.

## 2.2 Evaluation Indicators

Based on literature [8], multiple evaluation indicators are adopted as follows:

### 1) Efficiency Metrics

The first-law efficiency ( $\eta_1$ , energy efficiency) and second-law efficiency ( $\eta_2$ , exergy efficiency) serve as primary thermodynamic indicators, expressed as:

$$\eta_1 = \frac{W_{net}}{Q_{in}}$$

$$\eta_2 = \frac{W_{net}}{Ex_{in}}$$

where subscripts E and Ex represent energy and exergy values, respectively, and  $W_{net}$  denotes net power generation.

### 2) Primary Energy Saving Rate (PESR)

PESR evaluates the polygeneration system's performance relative to separate

production systems, defined as the energy savings achieved by the polygeneration system for the same product output:

$$\text{PESR} = [\text{MATH\_ERROR}]$$

where  $[\text{MATH\_ERROR}]$  represents the energy required by separate production systems for equivalent output, and  $\text{PE}$  is the system's energy input.

In this study, coal is assumed as the primary energy input, with the reference system comprising separate coal-to-SNG and coal-fired power generation systems. Biomass energy input is converted to equivalent coal energy input using a conversion factor ( $x$ ), representing the ratio of biomass quality to coal quality [9]:

$$\text{PE} = [\text{MATH\_ERROR}]$$

---

### 3.1 Design Condition Thermodynamic Analysis

The novelty of the proposed system lies in the biomass-coal complementarity that promotes the methanation process and the CLC unit that achieves zero-energy-penalty CO capture. Table 3 presents the thermodynamic performance of the new system, while Table 4 shows the primary energy saving rate.

As shown in Tables 3 and 4, under the specified conditions with 40 wt.% of coal replaced by biomass, the polygeneration system achieves a first-law efficiency ( ) of 57.03% and a second-law efficiency ( ) of 54.65%. The primary energy saving rate reaches 18.6% compared to separate production systems. These results demonstrate that the biomass-coal hybridization upgrades low-quality biomass energy to high-quality syngas while modifying the H /CO ratio to favor SNG synthesis.

---

### 3.2 Parameter Analysis

The main parameters affecting the polygeneration system include the biomass-to-coal complementarity ratio, oxygen-to-carbon ratio (O/C), steam-to-carbon ratio (S/C), and unreacted gas recycle ratio. The biomass-coal complementarity advantage stems from coal's high carbon content and biomass's high hydrogen content, where different blending ratios influence the syngas H /CO ratio and consequently affect the methanation process. The O/C and S/C ratios influence gasification equilibrium, thereby regulating the H /CO ratio. The unreacted gas recycle ratio directly impacts reactant conversion and methane yield, thus affecting overall system performance. This section examines the influence of these four parameters.

#### (1) Effect of O/C Ratio on System Performance

The O/C ratio determines the carbon conversion efficiency in solid fuel gasifica-

tion. [Figure 2: see original paper] shows the effect of O/C ratio and biomass-coal complementarity ratio on syngas yield. The results indicate that syngas yield reaches its maximum at O/C = 0.4 for all biomass blending ratios from 10% to 40%. Additionally, syngas yield decreases slightly with increasing biomass proportion due to biomass' s lower carbon content compared to coal, which reduces CO generation.

## (2) Effect of S/C Ratio on System Performance

[Figure 3: see original paper] illustrates the influence of S/C ratio on syngas H /CO ratio adjustment and exergy destruction in the first-stage SNG reactor. Increasing the S/C ratio improves the H /CO ratio by promoting the water-gas shift reaction, which enhances H production while consuming CO. However, higher S/C ratios also increase exergy destruction in the first-stage SNG reactor because the additional CO generated from the shift reaction is detrimental to methanation.

When biomass replaces 部分 coal feedstock, the syngas H /CO ratio increases with the biomass-coal complementarity ratio. For instance, at S/C = 0.4, the H /CO ratio rises from 0.69 (without biomass) to 0.83 (with 40% biomass blending). This leverages biomass' s higher hydrogen content and coal' s higher carbon content to optimize the syngas composition for methanation. Consequently, increased biomass proportion reduces the irreversible losses in the first-stage SNG reactor.

[Figure 4: see original paper] shows the combined effect of S/C ratio and biomass blending on system efficiency. Without biomass (B = 0), as S/C increases from 0.25 to 0.6, the overall energy efficiency ( ) increases from 57.40% to 60.37%, while exergy efficiency ( ) rises from 55.72% to 58.83%—an improvement of approximately 3 percentage points. However, system efficiency begins to decline when S/C exceeds 0.6. While higher S/C ratios improve the H /CO ratio and promote methanation (increasing SNG production), they also intensify steam consumption, reducing the steam available for power generation. When S/C surpasses 0.6, the SNG production increase cannot compensate for the power generation loss, causing efficiency to drop.

At a biomass blending ratio of 0.4, the efficiency trend with S/C remains similar but shifts to lower values, with optimal overall energy efficiency ( ) of 57.30% and exergy efficiency ( ) of 54.93%—approximately 3 percentage points lower than the optimal case without biomass ( = 60.37%). Notably, the optimal S/C value decreases from 0.6 (without biomass) to 0.4 (with 40% biomass), indicating reduced water consumption. This demonstrates that biomass-coal complementarity not only improves the H /CO ratio and reduces methanation irreversibility but also leverages biomass' s inherent moisture content to decrease process water demand.

## (3) Effect of Unreacted Gas Recycle Ratio on System Performance

The unreacted gas recycle ratio (RR) is defined as the mass flow ratio of recycled unreacted gas to fresh feed gas. [Figure 5: see original paper] shows

RR' s impact on exergy destruction in the first-stage SNG reactor. Without biomass feedstock, as  $RR/(RR+1)$  increases to approximately 0.8, exergy losses in the first-stage SNG reactor increase gradually but remain limited. Beyond this “inflection point,” further increasing RR causes a sharp rise in exergy destruction. Although SNG output continues to increase beyond the inflection point, the marginal gain is minimal. This phenomenon reveals that the conventional “exhaustive conversion” approach—maximizing feedstock conversion and methane yield through high recycle ratios—incur substantial energy penalties, where enormous energy losses yield only modest SNG production increases.

As the biomass-coal complementarity ratio increases from 0 to 0.4, the trend of SNG reactor exergy destruction with RR becomes progressively flatter, with the inflection point becoming less pronounced. The biomass-coal hybrid effectively improves the syngas  $H_2/CO$  ratio, reducing methanation difficulty and thereby decreasing reactor irreversibility. Consequently, exergy destruction decreases with higher biomass proportions, and the inflection point phenomenon diminishes.

[Figure 6: see original paper] illustrates the effect of RR on overall system efficiency and PESR. Without biomass ( $B = 0$ ), both system efficiency and PESR initially increase then decrease with  $RR/(RR+1)$ . While SNG production increases with RR, system efficiency begins declining when RR exceeds 0.7 due to escalating compression power requirements. PESR decreases continuously with  $RR/(RR+1)$ , albeit modestly, primarily because increased recycle power reduces net power generation in the polygeneration system. Biomass feedstock (40% blending) reduces system efficiency by approximately 3 percentage points, attributable to the difference in heating values between biomass and coal. However, biomass significantly enhances PESR—at a 40% blending ratio, PESR improves by about 4 percentage points compared to the no-biomass case—demonstrating that biomass-coal complementarity upgrades biomass energy quality to syngas level for SNG synthesis and power generation.

---

## 4 Conclusion

To resolve the issues of inefficient energy utilization and high  $CO_2$  capture energy penalties in conventional coal-to-SNG processes, this paper proposes a hybrid biomass-coal polygeneration system integrated with chemical looping combustion. Under standard conditions, the system achieves an overall energy efficiency ( ) of 57.03%, exergy efficiency ( ) of 54.65%, and a primary energy saving rate of 18.6%, while realizing zero net  $CO_2$  emissions. The biomass-coal complementarity enhances the syngas  $H_2/CO$  ratio, promoting methanation and thereby reducing process irreversibility, water consumption, and improving primary energy savings.

## References

- [1] HUANG Pinghui, ZHANG Shuying, QIN Qirong, et al. The situation, demand and countermeasures for natural gas resources in China [J]. Xinjiang Petroleum Geology, 2005, 26(1):105-107.
- [2] HAN Jingkuan, ZHOU Shuhui, TIAN Ying, et al. Prospect of coal-based synthetic natural gas (SNG) projects from the perspective of market supply and demand [J]. Natural Gas Industry, 2014, 34(7):115-122.
- [3] LI Sheng. The mechanism of minimal energy penalty for CO capture and the study on coal-based polygeneration system for cogenerating substitute natural gas and power [D]. Beijing: Chinese Academy of Sciences (Institute of Engineering Thermophysics), 2012.
- [4] Muresan M, Cormos C C, Agachi P S. Techno-economical assessment of coal and biomass gasification-based hydrogen production supply chain system[J]. Chemical Engineering Research & Design, 2013, 91(8):1527-1541.
- [5] Larson E D, Fiorese G, Liu G, et al. Co-production of decarbonized synfuels and electricity from coal + biomass with CO capture[J]. Energy & Environmental Science, 2010, 3(1):28-42.
- [6] Abdelouahed L, Authier O, Mauviel G, et al. Detailed modeling of biomass gasification in dual fluidized bed reactors under Aspen Plus[J]. Energy & Fuels, 2012, 26(6):3840-3855.
- [7] Porrazzo R, White G, Ocone R. Techno-economic investigation of a chemical looping combustion based power plant[J]. Faraday Discussions, 2016, 192.
- [8] Li S, Jin H, Gao L. Cogeneration of substitute natural gas and power from coal by moderate recycle of the chemical unconverted gas[J]. Energy, 2013, 55(55):658-667.
- [9] Lin H, Jin H, Gao L, et al. A polygeneration system for methanol and power production based on coke oven gas and coal gas with CO recovery[J]. Energy, 2014, 74(2):174-180.

### Corresponding Author:

HONG Hui, Institute of Engineering Thermophysics, Chinese Academy of Sciences, No. 11 Beisihuan West Road, Haidian District, Beijing 100190, China. Tel: 18701009257, Email: fanjunming@iet.cn

*Note: Figure translations are in progress. See original paper for figures.*

*Source: ChinaXiv – Machine translation. Verify with original.*