

The high efficient synthesis of natural gas from a joint-feedstock of coke-oven gas and pulverized coke via a chemical looping combustion scheme



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HIGHLIGHTS

- Proposed a new process of pulverized coke CLC-assisted COG for producing SNG.
- Improved hydrogen utilization efficiency and SNG capacity by 20% in the new process.
- The co-feeding process resulted in exergy saving by 6.8% with IRR increase of 2.3%.

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ABSTRACT

Natural gas is regarded as a high heating-value and low carbon-footprint fuel, but its supply is deficient and global imbalanced. A technical routine toward synthetic natural gas (SNG), sound in both energy efficiency and environmental impact, is therefore highly demanded. Both coke-oven gas (COG) and pulverized coke (PC) are two major by-products of metallurgical industries. There is the conventional SNG process fed by only COG (CGtSNG), which suffers from less hydrogen utilization and energy efficiency, as well as limited SNG capacity. The driving force on the environmentally-responsible and energy-efficient use of the PC calls for the involvement of state-of-the-art chemical looping combustion (CLC) technology. A new process co-fed by both PC and COG for SNG production via a CLC unit (PC_{CLC}-CGtSNG) is therefore proposed in this study. The PC_{CLC}-derived CO₂ can be an effective carbon source to optimize the composition of syngas for high efficient SNG production. The involvement of the CLC in the conventional CGtSNG can achieve intensive material and energy couplings, improve energy and hydrogen utilization efficiencies, and increase SNG productivity. Process modelling results showed that the proposed process possessed about 20% improvement in the hydrogen utilization efficiency and SNG productivity, as well as 6.8% increase of the exergy efficiency, comparing to those of the conventional CGtSNG.

1. Introduction

Natural gas is a clean energy and resource with high heating value and low carbon-footprint. It is a promising candidate replacing coal to reduce air pollution emissions, especially under the current carbon emission constraints. However, the supply of natural gas is deficient and global imbalanced. Taking an example of China, its production was only $1.38 \times 10^{11} \text{ m}^3$ in 2016, but the natural gas demand was as high as $2.10 \times 10^{11} \text{ m}^3$ over the same period [1]. There is the conventional coke oven gas (COG)-to-synthetic natural gas (SNG) process (CGtSNG) [2]. However, it suffers from less hydrogen utilization efficiency and

energy efficiency, as well as limited SNG capacity. The ideal molar ratio of hydrogen to carbon in syngas, defined as an R value, ($R = (\text{H}_2 - \text{CO}_2)/(\text{CO} + \text{CO}_2)$, on the basis of molar), is about 3 for SNG production [3,4]. For the carbon-rich fuels of coal and coke, the water gas shift reaction is generally involved to increase hydrogen concentration of syngas for efficient SNG production. Therefore, the coal or coke-to-SNG processes suffer from serious CO₂ emissions [5].

Both COG and pulverized coke (PC) are two major by-products of metallurgical industries. The COG is mainly composed of enriched H₂ (58–60 vol.%), CH₄ (23–27 vol.%), CO (5–8 vol.%), and CO₂ (< 3 vol.%) [6]. The CGtSNG is therefore promising, especially contrasting to

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Nomenclature

| | |
|------------|---|
| η | exergy efficiency |
| φ | hydrogen utilization |
| R | molar ratio of $(H_2-CO_2)/(CO + CO_2)$ |
| ΔG | gibbs free energy change |
| GT | gas turbine |
| OC | oxygen carrier |
| PC | pulverized coke |
| MU | methanation unit |
| CLC | chemical looping combustion |
| COG | coke-oven gas |

| | |
|-------------------|----------------------------------|
| FCI | fixed capital investment |
| HPS | high pressure steam |
| HRU | heat recovery unit |
| IPS | intermediate pressure steam |
| IRR | internal rate of return |
| LPS | low pressure steam |
| SNG | synthetic natural gas |
| TPC | total product cost |
| AGRU | acid gas removal unit |
| CLCU | chemical looping combustion unit |
| CGtSNG | coke-oven gas-to-SNG |
| $PC_{CLC-CGtSNG}$ | PC CLC-assisted COG-to-SNG |

the direct burning the COG. The CO and CO₂ of the COG is reacted with H₂ to generate SNG in the CGtSNG, which reduces the difficulty of separating H₂ and CH₄. The ideal R is about 3 for SNG production; however, the R ratio of the COG feedstock is theoretically about 5.2 [7]. The hydrogen wasting is unavoidable in the CGtSNG and the carbon input is therefore demanded for the improvement of the process.

Combining coal/coke-derived syngas and COG is therefore a rational approach to realize an ideal R ratio for SNG production. Many studies focused on the techno-economic analysis and system optimization on cogeneration of SNG and power based on coal gasification and/or COG reforming. For example, the overall energy efficiency of the polygeneration plant can reach 62%, much higher than only 55% in a coal-to-SNG plant [8,9]. Man et al. [10] combined coal gasification and COG dry reforming to adjust the R ratio of syngas for SNG production. The energy efficiency of the co-feeding system was increased by 4% and CO₂ emissions was reduced by 60%. Besides, various ways for producing different chemicals such as methanol, dimethyl ether, and olefins

by co-feeding of coal and COG had been proposed in recent years [11–13]. Above research was focused on improving techno-economic performance of the coal derived chemicals processes. However, the research on techno-economic improvement of the COG derived chemicals processes is rare. Yi et al. [2] proposed a new process of CO₂ recycle assisting the CGtSNG, improving the energy efficiency by 6.6%, reducing CO₂ by 99.9%, and increasing the SNG output by 20%. Similar works also included the CO₂ dry reforming of the COG for methanol production [6,7,14,15]. However, the aforementioned processes incurred the supply of costly pure CO₂ separated from tail gas or oxygen-enriched combustion gas, in which significant energy penalty was inevitable.

Chemical looping combustion (CLC) can efficiently exclude costly separation of air and provide the pure CO₂ [16,17]. The CLC uses solid oxygen carrier (OC) to oxidize fuels in a fuel reactor (FR) and solid OC is recycled by the oxidization using air in an air reactor (AR) [18,19]. The primarily product of FR are CO₂ and H₂O, and a high concentrated

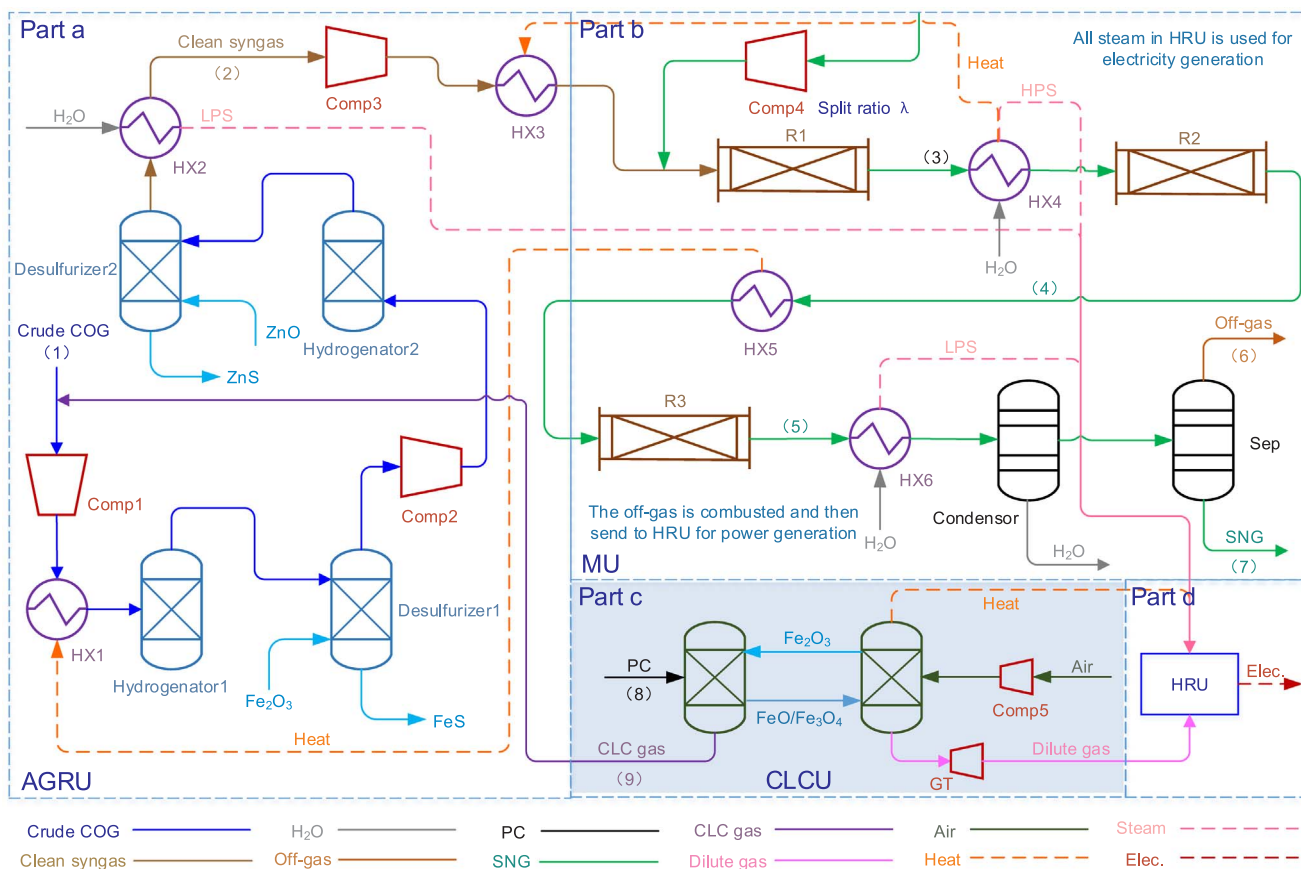


Fig. 1. A concept diagram of the PC_{CLC}-CGtSNG.

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