



Thermodynamic assessment of SNG and power polygeneration with the goal of zero CO₂ emission

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ARTICLE INFO

Article history:

Received 10 October 2017

Received in revised form

20 January 2018

Accepted 9 February 2018

Available online 12 February 2018

Keywords:

Polygeneration

Chemical looping combustion

SNG

CO₂ capture

Energy and economic analysis

ABSTRACT

The current coal-to-SNG (synthetic natural gas) projects are facing two main issues, addressing on insufficient energy utilization and large amount of carbon emissions. For this reason, we proposed a coal-based SNG and power cogeneration process, at the ability to achieve high-efficiency and achieve zero-energy-penalty separation of CO₂, by incorporation with chemical looping combustion (CLC). The main objective of this paper is to estimate thermodynamic performances under the effects of key parameters in order to pre-examine whether the proposed process is thermodynamically viable. At optimal condition (oxygen to carbon ratio (O/C) = 0.4, steam to carbon ratio (S/C) = 0.8 and recycling ratio of unreacted gas (Ru) = 3), the overall plant energy efficiency, exergy efficiency and energy saving ratio are 58.84%, 56.59% and 10.82%, respectively. The proposed process is compared with several traditional systems that aims to further demonstrate the superiority of such process design in terms of thermodynamic and preliminarily economic assessment. A sensitivity analysis is also included to examine the effects of different coal feedstocks and oxygen carriers of CLC on system performances.

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1. Introduction

Coal-to-SNG technology has attracted considerable attentions in recent years. Especially in China, it represents as one promising technology to achieve clean coal utilization and address energy crisis by continuously producing natural gas from abundant reserve of coal. Particularly, this sense has been realized by Chinese policy-maker that helps to ensure energy security, reduce natural gas imports, stabilize fuel price and diversify energy options [1]. Therefore, the Chinese government has vigorously promoted coal-to-SNG projects; several projects have been planned and under constructed already being demonstrated with two representative plants, i.e. Shenhua Corporation 2 billion cubic meters per year in Inner Mongolia and Datang Corporation 4 billion cubic meters per year in Liaoning [2].

The research associated with coal to SNG technologies mainly focuses on the following aspects: (1) The design of methanation reactor and synthesis catalysts of SNG were reported in Refs. [3,4]. (2) Life cycle analysis of coal to SNG system can be found in Refs. [5,6]. (3) The economic assessment of coal to SNG system was

reported in Refs. [7,8]. In addition, a comprehensive review of SNG technology from coal has been summarized by Jan et al. [9] in 2010 and Xu et al. [10] in 2015 as well as the recent review carried out by Rönsch et al. [11] in 2016.

However, in traditional coal-to-SNG plant, the by-product power is generated by recovering exothermic methanation reactions that generally cannot offset the inherent work requirement (e.g. compression work) [12]. In addition, in order to maximize reactions conversion, it primarily requires the favored stoichiometric synthetic gas composition ($H_2/CO = 3:1$) and enforces unlimited recycle ratio of unreacted gas. However, the former one can lead to considerable exergy destructions [13]; unlimited recycle ratio of unreacted gas enhances the energy penalty burden for chemical synthesis [14].

The SNG and power polygeneration is at present one of the most promising concept to address both issues. Three major achievements have been outlined. First, the polygeneration system eliminates water-gas shift process being demonstrated with higher energy efficiency without WGS adjustment that simultaneously reduces fresh water consumption [12]. Secondly, reasonably recycling unreacted gas to methanation unit controls remarkable exergy destruction within methanation that simultaneously enables by-product power generation from unreacted gas, providing

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better utilization of coal energy [5]. Finally, enhancement of CO₂ concentration provides less energy penalty for carbon mitigation [7].

Facing the effective approach to reducing carbon footprint represents another barrier for such polygeneration process development with the increasing restriction on greenhouse gas emissions. Carbon dioxide capture and storage (CCS) can be utilized in coal chemistry industry to help significant CO₂ emission reductions. However retorting CCS in coal-to-SNG plants is difficult and currently is not feasible because of the huge energy penalty on carbon mitigation (e.g. carbon capture by amine scrubbing reduces the net efficiency by approximately 15% [15]) and high investment costs.

Fortunately, chemical looping combustion (CLC) is posed to be one of the most promising options as a long-term implementation for carbon capture. CO₂ is inherently separated and very few energy penalty is expected for carbon capture [16]. Compared with traditional fuel direct combustion, CLC decomposes direct combustion reaction into two sub-reactions, which are correspondingly occurred in a fuel reactor (FR) and an air reactor (AR) [17]. In the FR, the fuel gas or syngas is almost completely converted into H₂O and CO₂ (restricted by the thermodynamic limitation), while the oxygen carrier (e.g. NiO) is reduced to its reduced state (Ni). Moreover, the reduced oxygen carrier (Ni) is transported back to the AR and is reacted with air to regenerate its original state (NiO). At this stage, without dilution of N₂, pure CO₂ can be easily obtained through water vapor condensation [18].

Inspired by this, integrating CLC into coal-to-SNG and power polygeneration system might be very attractive for the goal of zero-energy-penalty carbon capture by burning the unreacted gas from methanation unit. This is the aim of this paper which examines the potential benefits and technical feasibility of such process from both energy-efficient and environmental-sound views. The novelties of this polygeneration system are shown below:

- (i) This design provides a novel method for carbon capture by using CLC in polygeneration system. The function of CLC integrated into this polygeneration system aims for CO₂ concentration enrichment during combustion at the demand for inherent carbon capture. Therefore we intend to highlight the interaction between thermodynamic performance of such processes and the implement of CLC. For such purpose, we have examined the effects of several essential parameters that have not been deeply analyzed in previous literature, such as steam to carbon ratio (S/C), oxygen to carbon ratio (O/C), types of oxygen carriers, etc.
- (ii) Besides, some may concern the introduction of CLC in polygeneration system would increase the cost of SNG yield, we therefore perform an economic assessment of this newly designed process and intend to present its potential economic benefits to the community as well as to the policy-marker.

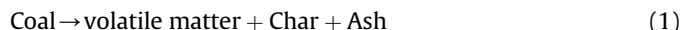
2. Process description

The general process considered in this study allows the polygeneration of SNG and power with simultaneously carbon capture, as shown in Fig. 1. It consists of three major sections: gasification and purification system, SNG system, and CLC system.

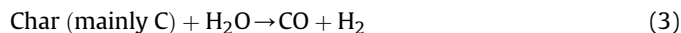
2.1. Gasification and purification system

The coal gasification is a complicated process involving a series of physical and chemical reactions. Initially, the coal is thermally

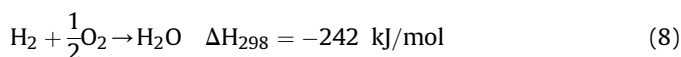
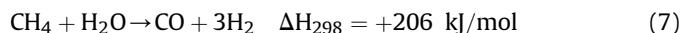
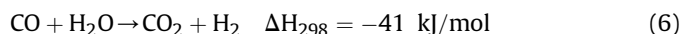
devolatilized with a rapid pyrolysis, shown below [19]:



The volatile matter mainly consists of H₂, CO, CO₂, CH₄ and other undesirable gas, such as H₂S, COS, NO_x and other light hydrocarbons. Moreover, the solid char particles can react with steam, carbon dioxide and oxygen to produce syngas, shown as follows [20]:



In addition, water-gas shift (WGS) reaction, steam-methane reforming reaction and H₂ partial combustion reaction are also included [21]:



Within gasification process coal and oxidants (oxygen, steam) react in a gasifier to produce syngas, where the necessary oxygen and steam are derived from ASU (air separate unit) and downstream Heat Recovery Steam Generator & Steam Turbine (HRSG & ST) system, respectively. Associated with the definition of coal as unconventional component in Aspen plus, RYield model is used to simulate coal decomposition. In this step, coal is converted into its basic constituting components (i.e. H₂O, O₂, C, S and Ash). Subsequently, RGibbs model is employed to simulate coal pyrolysis and gasification process that is based on chemical equilibrium restricted by minimizing Gibbs free energy subject to restrict the thermodynamic limitation [22]. The excessive heat from syngas can be recovered in a waste heat boiler (WHB) to generate steam (540 °C and 120 bar). In order to obtain high-quality syngas that avoids downstream methanation catalyst from devastation, the cooled syngas is sent to the purification unit to remove ash and desulfurization. Ash is separated by the cyclone separator and sulfur compounds are removed by the Selexol gas clean technology [23].

In order to verify the accuracy of the gasification model, simulation results are compared with the experiment data given in the literature [24] under the consistent gasification conditions (1346 °C and 28 bar), shown in Fig. 2. This closed results reveal that simulation results are reliable within tolerant errors.

2.2. SNG system

The flow diagram of SNG unit being adopted from TREMP technology is shown in Fig. 3. To convert syngas into SNG and effectively remove excessive heat, three series adiabatic reactors with intercooling are needed. Notably, for improving syngas conversion, a water-gas shift (WGS) reaction is placed before methanation unit by adjusting H₂/CO in syngas to stoichiometric ratio of 3 in conventional coal-to-SNG plant [25]. Despite of its potential benefit for methanation, it increases the exergy losses of the subsystems that is not beneficial to overall system performance [12,13]. Therefore WGS process is rejected from consideration in this study.

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