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Integrated process for synthetic natural gas production from coal and coke-oven gas with high energy efficiency and low emission

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ABSTRACT

There was a rapid development of coal to synthetic natural gas (SNG) projects in the last few years in China. The research from our previous work and some other researchers have found coal based SNG production process has the problems of environmental pollution and emission transfer, including CO_2 emission, effluent discharge, and high energy consumption. This paper proposes a novel co-feed process of coal and coke-oven gas to SNG process by using a dry methane reforming unit to reduce CO_2 emissions, more hydrogen elements are introduced to improve resource efficiency. It is shown that the energy efficiency of the co-feed process increases by 4%, CO_2 emission and effluent discharge is reduced by 60% and 72%, whereas the production cost decreases by 16.7%, in comparison to the conventional coal to SNG process. As coke-oven gas is a waste gas in most of the coking plant, this process also allows to optimize the allocation of resources.

1. Introduction

In China, coal accounts for 67.4% of the total energy consumption [1]. Too much coal consumption, insufficiently clean processing and burning have brought about serious haze problem in China. Using natural gas to replace coal is one of the key measures to tackle with the air pollution caused by coal combustion [2]. However, the production of natural gas is only 1.57×10^{11} m^3 (including the production of shale gas) while the natural gas consumption is about 2.5×10^{11} m^3 in 2015 [3]. China is facing the problem of supply shortage of natural gas.

Aiming at the promising market opportunity, Chinese industry made a great efforts to push forward coal to synthetic natural gas (CtSNG) process in recent years. Till now, there have been 8.73×10^{10} m^3/y production capacity of CtSNG projects in commercial operation in China [4].

A schematic diagram of CtSNG process is shown in Fig. 1. Feedstock raw coal is gasified to crude syngas. The crude syngas is sent into the water gas shift (WGS) unit to adjust the hydrogen-to-carbon (H/C) ratio. Next the syngas is sent into the acid gas removal (AGR) unit to remove CO_2 and sulfide. Finally, the clean syngas is sent into methanation unit for producing SNG [5]. Li et al. [6] made a comparison of techno-economic and environmental performance between coal-based SNG and coal directly burning. The results showed that the coal based SNG using in residential sector could relieve urban haze pollution caused by the direct coal burning. However, coal based SNG has nearly two times higher consumption of raw material and 1.65 times higher CO_2 emission than coal burning.

CtSNG process uses lignite as the feedstock due to the limitation of resource allocation. That means the crude syngas from gasification would contain large amount of CO_2. H/C ratio of the crude syngas is around 2.3–2.7, while the required H/C ratio for methanation is about 3.1–3.3. The syngas is lack of hydrogen source. CO of the syngas needs to be converted to H_2 in WGS unit. It results in a high emission of CO_2 and waste of carbon resource, as well as very high consumption of energy in the WGS unit. Meanwhile, the crude syngas from Lurgi gasification needs to be washed with water to remove the phenol and ammonia. This process generates almost 98% waste water of the CtSNG process [7]. To treat the effluent with high concentration of phenol and ammonia would consume large amount of energy together with high production cost. Discharge of the effluent without fully treatment would cause serious impact to the environment [8].

Coke-oven gas (COG) is generated in the coking process as a by-product of coal carbonization. The annual COG production is estimated at 7×10^{10} m^3 in China [9]. However, most of the COG is directly discharged into the atmosphere. It causes considerable energy waste and serious environmental consequences [10]. COG is a kind of hydrogen-rich gas, which contains H_2 (55–60%), CH_4

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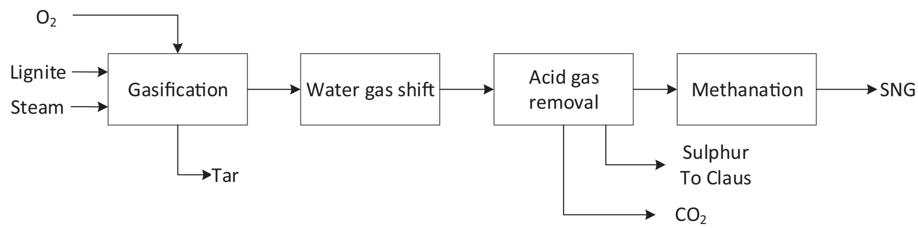

Fig. 1. Schematic diagram of CtSNG.

(23–27%), CO (5–8%), N_2 (3–6%), CO_2 (less than 2%). Co-feed system could take advantage of coal and COG to produce syngas with suitable H/C ratio. Authors' previous work [11] on COG assisted coal to olefins process found that the energy efficiency of co-feed system improved by 10% and the CO_2 emission decreased by 85% when compared with single coal feed. The co-feed process of coal and COG to SNG is a promising alternatives in dealing with the problems of high CO_2 emission and energy consumption. In China, most of CtSNG projects are surrounded by coal coking plants. Plentiful COG source makes assurance for feedstock supply for the co-feed system of coal and COG to SNG process.

Following the above ideas, this paper proposes a novel co-feed process of coal and COG to SNG with high energy efficiency and low CO_2 emission. The co-feed process integrates a dry methane reforming unit to reduce CO_2 emission, and uses the hydrogen resource in COG to raise the H/C ratio of the syngas. A conceptual design and a techno-economic analysis for the co-feed process is put forward in this paper.

2. Coal and coke-oven gas to SNG process

The schematic diagram of the proposed co-feed process of coal and COG to produce SNG (CGtSNG) is shown in Fig. 2. CGtSNG process contains coal gasification unit, acid gas removal unit, and methanation unit. However, it removes the WGS unit, while adds a COG separation unit and a dry methane reforming (DMR) unit compared with CtSNG process. Part of methane from COG is sent into DMR unit to react with CO_2 from the coal gasification unit. DMR unit produces clean syngas and reduces CO_2 emission. The syngas with suitable H/C ratio for methanation is obtained by adjusting the proportion of DMR syngas, coal gasification syngas, and H_2 from COG.

CGtSNG process is composed of three subsystems: (1) coal gasification and syngas clean; (2) COG separation and dry methane reforming; (3) methanation. The subsystems are introduced as follows. Feedstock composition of CGtSNG is shown in Table 1. The proximate and ultimate analysis of lignite is shown in Table 2. The data is referred to the lignite from Shengli coalfield of Xilinhot, China. To facilitate comparison between CGtSNG and CtSNG, the production capacity of SNG in CGtSNG is set to 4.0 billion m^3/y, same as industrial CtSNG project of Datang Corporation in Chifeng, China [7].

2.1. Coal gasification and syngas clean

Due to the resource allocation limitation, all the coal to SNG processes in China use lignite as the raw material. That means only Lurgi and BGL gasification technologies could be used in CtSNG. The syngas from Lurgi gasification contains 39% H_2, 14% CO, and 32% CO_2. The syngas from BGL gasification contains 28% H_2, 55% CO, and 6% CO_2. BGL has advantage in low CO_2 content. However, H/C ratio of BGL gasification syngas is as low as 0.5, while the H/C ratio of Lurgi gasification syngas is 2.7. For methanation, the H/C ratio is required above 3.05 at least. That means a large scale of water gas shift unit is needed to convert CO into CO_2 and H_2 to raise the H/C ratio. The total amount CO_2 for CtSNG process of BGL is 18.8% higher than that of Lurgi. That also means that the handling scale of coal gasification unit and acid gas removal unit should be enlarge which causes the increasing of capital investment and operation cost. In China, the existent CtSNG projects in commercial operation uses Lurgi gasification technology.

The subsystem of coal gasification and syngas clean selects Lurgi pressurized coal gasification technology and Rectisol methanol washing technology as the simulation model referred to the CtSNG project of Datang Corparation in Chifeng, China. The flowsheet of this subsystem is shown in Fig. 3.

In coal gasification process, lignite is sent into gasifier together with oxygen from air separation unit (ASU) and steam. This process begins with lignite drying and pyrolysis, and continues with combustion and steam gasification. Main reactions in gasification

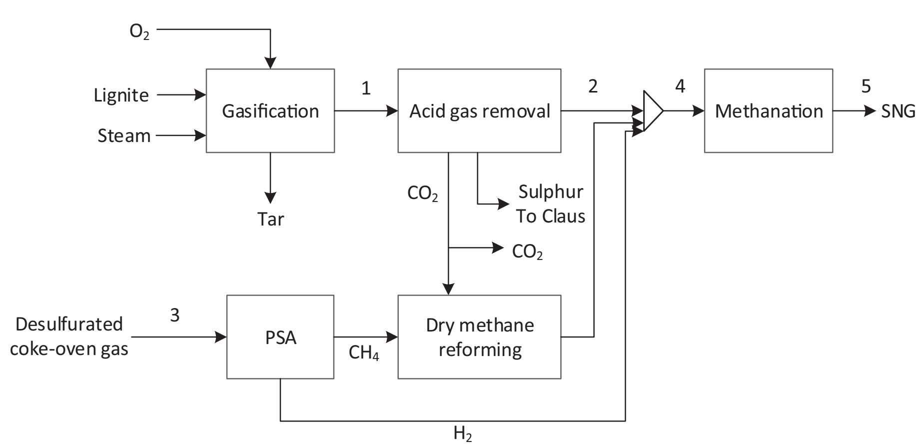

Fig. 2. Proposed schematic diagram of CGtSNG process.