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Performance analysis of solar energy integrated with natural-gas-to-methanol process

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ABSTRACT

Methanol is an important platform chemical. Methanol production using natural gas as raw material has short processing route and well developed equipment and technology. However, natural gas reserves are not large in China. Solar energy power generation system integrated with natural-gas-to-methanol (NGTM) process is developed, which may provide a technical routine for methanol production in the future. The solar energy power generation produces electricity for reforming unit and system consumption in solar energy integrated natural-gas-to-methanol system (SGTM). Performance analysis of conventional natural-gas-to-methanol process and solar energy integrated with natural-gas-to-methanol process are presented based on simulation results. Performance analysis was conducted considering carbon efficiency, production cost, solar energy price, natural gas price, and carbon tax. Results indicate that solar energy integrated with natural-gas-to-methanol process is able to cut down the greenhouse gas (GHG) emission. In addition, solar energy can replace natural gas as fuel. This can reduce the consumption of natural gas, which equals to 9.2% of the total consumed natural gas. However, it is not economical considering the current technology readiness level, compared with conventional natural-gas-to-methanol process.

1. Introduction

Methanol is an important platform chemical [1]. There are hundreds of chemical products, which can be synthesized from methanol. The current annual methanol production capacity exceeds 31 million tons in China [2]. From 2003 to 2014, the methanol production has increased significantly. Methanol production in 2014 was nearly 10 times higher than that in 2003, which is shown in Fig. 1 [3]. At the same time, consumption of methanol reached 33 million tons in 2014, accounting for nearly 40% of the global total demand of 65 million tons. It is estimated that annual methanol production capacity will exceed 66 million tons of methanol by 2020 in China [4].

Natural-gas-to-methanol is a way to produce methanol [6]. Most of the methanol production facilities outside China are using natural gas as raw material. The methanol production using natural gas or low carbon hydrocarbon takes about 70% of the total methanol production capacity [7]. The natural gas route methanol plant is mainly distributed in oil fields or natural gas producing areas in China, such as the Northwest Sichuan methanol plant, Golmud Vico methanol plant, Daqing methanol plant, Changqing methanol plant, CNOOC 600 thousand tons of methanol, etc. A few plants use natural gas from the west east gas pipeline to produce methanol, for example, CNOOC, tiye chemical 200 thousand tons methanol plant and Henan Zhumadian 300 thousand tons methanol device [8].

Methanol production using natural gas as raw material has short process route and well developed equipment and technology [6]. However, natural gas reserves are not large in China. There is still a big gap in the supply of natural gas market [8]. The cost of natural gas routes is relatively high [9]. Many researchers are constantly committed to the study of various NGTM processes [10,11]. Renewable energy integration is one of the methods [12,13]. Solar energy is one kinds of renewable energy. Zeng et al. experimentally tested the performance of solar energy integration to drive biomass pyrolysis [14]. Soares et al. developed a system that integrated solar energy to drive an Organic Rankine Cycle, the system annual yield is significantly increased 3.4–9.6% [15]. Tanaka et al. injected solar energy into the heat

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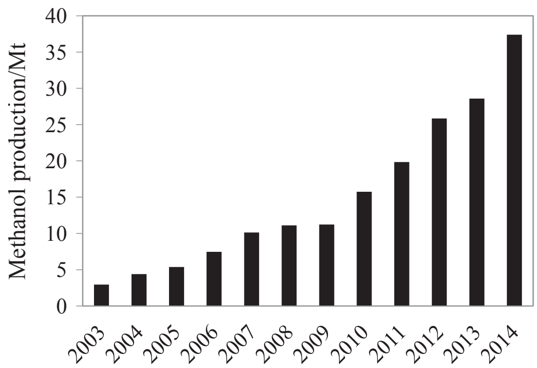

Fig. 1. Total methanol production of China from 2003 to 2014 [5].

recovery steam generator (HRSG) combined cycle system [16]. Hou et al. developed a system to integrate solar energy where the solar energy was used to replace the extraction steam for preheating the feed water [17]. Peterseim et al. investigated that the net efficiency of the solar to electricity can be improved by biomass materials [18,19]. Soria et al. proposed a strategy for developing the solar power according to the renewable energies situation of Brazil [20]. Bai et al. proposed and evaluated solar energy integration system for producing the methanol and electricity [21]. Storch et al. developed and simulated that produces methanol via indirectly heated solar reforming of natural gas, the proposed system indicated a promising efficiency [22].

To the best knowledge of the authors, few study has been done in the case of performance analysis of solar energy integrated with natural-gas-to-methanol process. Therefore, in this paper, solar energy integrated with natural-gas-to-methanol process is proposed. The performance of proposed system is evaluated, including carbon efficiency, production cost, natural gas price, solar energy price, and carbon tax. The SGTM provides a way for the utilization of the abundant renewable solar energy resources and a promising solution to slow down GHG emission. In addition, solar energy can replace natural gas as fuel. This can reduce the consumption of natural gas.

2. Process simulation

2.1. Process description

Regardless of the adopted special techniques, generally, the whole process of NGTM consists of four generic subsystems, including an ASU (air separation unit), sulfur removal, natural gas reforming, and methanol synthesis. General plant support includes power generation, water treatment, and cooling tower. The process diagram is shown in Fig. 2. Natural gas is firstly compressed and heated, and then fed to sulfur removal reactor. A primary reformer and an autothermal reformer are employed to adjust the ratio of H_2 to CO to meet the requirement of the methanol synthesis. The sulfur removed natural gas is mixed with steam, and the mixture is compressed and reacted in primary reformer. A mixture mainly composed of CO, H_2, and CH_4, with O_2 generated from ASU is fed to autothermal reformer. The adjusted syngas is then sent into methanol synthesis to produce methanol.

2.1.1. Air separation unit (ASU)

ASU provides high purity O_2 for the gasification unit. In this work, cryogenic air separation is employed, among other options, due to the maturity of the process applied for large scale plants. During the process, air is firstly compressed and cleaned by the molecule sieve. It is then fed into a separation tower, which separates O_2 from N_2 and Ar. O_2 (95 mol%) is used as an oxidant in the autothermal reformer.

2.1.2. Sulfur removal unit

Sulfur removal unit is used to remove sulfurs. Inorganic sulfur is moved under the catalyst of iron oxide and organic sulfur is moved under the catalyst of activated carbon. Hydrogen reacts with the sulfurs under the action of ZnO catalyst. After the desulfurization reactions, the total sulfur of natural gas is less than 30 ppb (vol%). For modeling of sulfur removal unit, desulfurization reactor is modeled by using the Sep model.

2.1.3. Natural gas reforming

Natural gas reforming is a major technical route for large-scale synthesis of syngas or hydrogen. Reforming methods include steam methane reforming (SMR), dry methane reforming (DMR), methane partial oxidation (MPO), autothermal reforming (ATR), etc [23–25]. The fundamental reactions of the reforming methods are shown in Eqs. (1)–(4).

$$\text{SMR:}CH_4 + H_2\,O \rightarrow CO + H_2 \quad \Delta\,H= +247.3\ \text{kJ/mol} \tag{1}$$

$$\text{DMR:}CH_4 + CO_2 \rightarrow 2CO + H_2 \Delta\,H= +206.3\ \text{kJ/mol} \tag{2}$$

$$\text{MPO:}CH_4 + 0.5O_2 \rightarrow CO + H_2 \quad \Delta\,H= -35.6\ \text{kJ/mol} \tag{3}$$

$$l\text{ATR: }CH_4 + H_2\,O \rightarrow CO + H_2 \quad \Delta\,H= +247.3\ \text{kJ/mol}$$
$$CH_4 + 05O_2 \rightarrow CO + H_2 \quad \Delta H = -35.6\ \text{kJ/mol} \tag{4}$$

NGTM includes primary reformer and autothermal reformer. SMR is employed in primary reformer. ATR is employed in autothermal reformer.

2.1.4. Methanol synthesis

Low pressure method is explored in this paper, which includes Davy method, Lurgi method and Topsøe method [26]. The former two are better developed techniques. The advantages of Davy method are that the structure of the synthesis tower is simple and the equipment is easy to be manufactured. In contrast, the synthesis tower of Lurgi method is more complex [27]. For the low one-way conversion rate, Davy methanol synthesis process has to increase circulation of the unconverted gas, in order to reduce the discharge of purge gas. Conversely, the one-way conversion rate of Lurgi method is much higher, leading to circulation rate less than half of Davy method. Thus, the power consumption, related equipment size, and one-time investment of Davy methanol synthesis are larger than those of Lurgi methanol synthesis [28]. The upper part of Topsøe methanol synthesis tower is equipped with a sophisticated mechanism to prevent synthesis gas axial flow due to catalyst shrinkage, which results in low efficiency of catalyst [29]. Lurgi methanol synthesis process was selected in this work.

In general, there are several major reversible reactions in the methanol synthesis reactor. Cu-Zn-Al catalyst was used for this reaction with its suitable temperature 240 °C and pressure 8.2 MPa [30,31]. The main reactions are shown in Eqs. (5) and (6).

$$CO + 2H_2 \rightarrow CH_3OH \quad \Delta\,H= -90.84\ \text{kJ/mol} \tag{5}$$

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \quad \Delta\,H= -49.57\ \text{kJ/mol} \tag{6}$$

The syngas is fed to methanol synthesis with the ratio H/C of about 2. Following the synthesis reaction, the unreacted syngas is separated out from the chemical products and recycled back to the methanol synthesis to increase the methanol production. After the synthesis reaction, methanol production is fed to separation columns for methanol purification. In this study, methanol purification is adopted in the three column distillation process.

2.2. Flowsheet simulation

NGTM process discussed in this study was simulated by the flow sheet simulation software Aspen Plus [9,31,32]. Peng-Rob equation of state (EOS) was selected as the thermodynamic method [30]. Natural gas of 53,000 Nm^3/h was selected as the feedstock with its heat value of 35.1 MJ/Nm^3. The flow rate was determined to match an annual