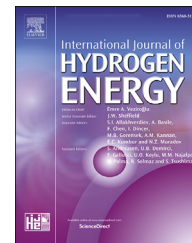




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# Hydrogen and LNG production from coke oven gas with multi-stage helium expansion refrigeration

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## ABSTRACT

Here we propose a novel cryogenic system to simultaneously produce liquid hydrogen (LH<sub>2</sub>) and liquefied natural gas (LNG) from coke oven gas. The coke oven gas, simplified as a mixture of methane and hydrogen, directly enters the cryogenic system. Due to the very low temperature of liquid hydrogen, helium is selected as the refrigerant, and the energy needed for the liquefaction is supplied by a multi-stage helium expansion refrigeration system. The high-purity liquid hydrogen and LNG products are obtained with the help of a cryogenic distillation column. The whole cryogenic process is simulated with the Aspen HYSYS software to determine the parameters of each process point and key component. We found that the process is able to produce LH<sub>2</sub> and LNG of very high purity. Using the power consumption of the product liquefaction as the major performance parameter for the analysis, optimum parameters of the multi-stage helium expansion liquefaction process could be found. The results show that the proposed system can achieve a methane recovery rate of 97.9% and a hydrogen recovery rate of 99.7% with acceptable energy consumption.

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## Introduction

Coke oven gas (COG) is a typical by-product in the coking process of coal [1]. China's coke production ranks highest in the world and reached 449 million tons in 2016. China produces 70 billion Nm<sup>3</sup> COG annually [2], and it is expected to remain important in the future as well [3]. Production of coke yields a lot of coke oven gas. Typically, one ton of coke generates approximately 360 Nm<sup>3</sup> of COG. The main components of COG are hydrogen (54–59%) and methane (24–28%) [4]. COG also contains small amounts of CO (5.5–7%), CO<sub>2</sub> (1–3%), N<sub>2</sub> (3–5%) and O<sub>2</sub> (0.3–0.7%) [4]. Most of the surplus COG is currently burnt off in torches and even in some cases directly emitted to the air [7,8]. Currently 20–40% of COG produced is normally utilized as fuel in the actual coke ovens [5–7]. Some

of the remaining COG generated is generally employed in alternative processes of the steel mills [1,2]. Consequently, COG represents serious ineffective use of energy, which also results in environmental pollution. COG can be regarded as a potential feedstock for hydrogen separation, methane enrichment, syngas and methanol production. Though the literature on utilization of COG and hydrogen or methane production from COG is ample [9–11], the literature on producing liquefied natural gas (LNG) and liquid hydrogen (LH<sub>2</sub>) simultaneously from COG is very scarce. Because both LNG and LH<sub>2</sub> are much more valuable than COG, a novel cryogenic system for liquid hydrogen and LNG production from coke oven gas is proposed in this paper.

There is a growing worldwide concern over the impact of climate change and pollution. Alternative clean energy fuels for the mobility and stationary power sector are an option to

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meet the targets for future greenhouse gas emissions and global warming, as indicated at the Paris Climate Conference in 2015 [12]. Hydrogen has shown great promise as an important energy source for use in the future. Liquid hydrogen has several advantages over other storage modes, especially in terms of energy density and convenience of utilization. Liquid hydrogen has been widely used in many situations [13–15]. Presently, liquid hydrogen is primarily used as a rocket fuel and is preferred for supersonic and hypersonic space vehicles mainly because of its small boiling point density and high unit mass. The wide ignition range in hydrogen/oxygen or air mixtures, as well as its large flame speed and cooling capacity due to its high specific heat, permits very effective engine cooling and cooling of the critical parts of the outer skin [14,16]. Liquid hydrogen has some other important uses such as in high energy nuclear physics and bubble chambers. Liquid hydrogen may also serve as transportation fuel, such as for fuel cell vehicles or hydrogen engine vehicles [17,18]. The transport of hydrogen is vastly more economical when it is liquefied even though cryogenic refrigeration and special dewar vessels are required. The recovery rate of liquefied hydrogen from COG for the cryogenic system proposed in this paper is over 98%. It can be seen that the system has great significance for the preparation and utilization of liquid hydrogen.

Hydrogen was firstly liquefied by a small device in 1898 [19]. Some years later, a pre-cooled Linde-Hampson system was used as the first simple laboratory system to liquefy hydrogen. Around 1900, more efficient laboratory systems were invented including the Claude, pre-cooled Claude, and helium-refrigerated systems, arranged in order to increase efficiency [20]. Next, in 1957, the first few large hydrogen plants were built in the USA for the growing petrochemical and aerospace industries and were based on the pre-cooled Claude cycle with more complicated systems that used liquid nitrogen as a pre-coolant to cool hydrogen gas down to  $-193\text{ }^{\circ}\text{C}$  and hydrogen refrigeration systems to further cool feed hydrogen gas to  $-253\text{ }^{\circ}\text{C}$  on a large scale [21].

Producing natural gas and/or hydrogen from coke oven gas is a new developing technology for comprehensive utilization of COG in recent years.

The methanization process [22] is a process for producing methane by reacting carbon monoxide with hydrogen. Coke oven gas is compressed after removing the benzene, naphthalene, heavy hydrocarbons and sulfides, and then carbon monoxide reacts with hydrogen to get the methane-based mixture gas. After that, the mixture gas goes into the pressure swing adsorption (PSA) device. Through the pressure swing adsorption separation technology, synthetic natural gas with methane more than 90% volume fraction will be produced.

The separation process [2] mainly includes the following steps. Firstly, pretreat the coke oven gas in order to remove the benzene, naphthalene, tar and HCN. Then proceed to the desulfurization, decarburization and drying processes. Then the coke oven gas flows along the hot side of a series of heat exchangers in which its temperature is lowered down. The cooled gaseous fluid enters into the gas-liquid separator, and the separated liquid is led into the cryogenic rectification column. The liquefied natural gas product will be obtained at the bottom of the cryogenic rectification column. Or omitting

the gas-liquid separator, introduce the gaseous fluid from the last heat exchanger directly into the cryogenic rectification column to distill it to get liquefied natural gas products.

Lin et al. [23] created and simulated a series of nitrogen expansion liquefaction processes for producing LNG from COG containing different amount of hydrogen at different methane recovery rates with Aspen HYSYS software. Analysis shows that, when the methane recovery rate is appropriate, the unit power consumption of the process with distillation is about  $0.7\text{ kWh/Nm}^3$ , and the methane content of LNG product is higher than 95%.

Onozaki et al. [24] proposed a technology for producing hydrogen from hot COG exhausted from coke ovens. The hot COG was introduced into an experimental unit, including a tar converter where oxygen and steam were injected. It was confirmed that most tar was partially oxidized and effectively converted to  $\text{H}_2$  and CO. The technology is promising, but sophisticated and costly.

Wang et al. [25] proposed alternative configurations for amplifying hydrogen from COG to improve COG energy conversion. The results show that integrating an upstream PSA separation with  $\text{CO}_2$  adsorption enhanced  $\text{H}_2$  amplification is an effective method for increasing the hydrogen yield and COG to  $\text{H}_2$  energy conversion efficiency. Detailed cost analysis was not included in the paper.

Bermúdez et al. [5] studied the  $\text{CO}_2$  reforming of coke oven gas for the production of synthesis gas over an activated carbon, an in-lab prepared  $\text{Ni/Al}_2\text{O}_3$  catalyst and physical mixtures of both materials in different proportions (AC + Ni) at  $800\text{ }^{\circ}\text{C}$ . Two different possible paths for the  $\text{CO}_2$  reforming of COG were researched.

The technologies mentioned above aim to product LNG or hydrogen from coke oven gas. However, the system proposed in this paper aims to produce both LNG and liquid hydrogen products through cryogenic methodology, which is novel but technically challenging.

## Comparison and selection of liquefaction system

### Comparison of LNG liquefaction processes

Currently, three main types of refrigeration cycle are used in LNG liquefaction processes, namely the cascade liquefaction process, the mixed refrigerant cycle liquefaction process and the liquefaction process with expander [26].

The cascade liquefaction process consists of three stages of independent vapor compression refrigeration processes, and each stage provides cooling capacity liquefying natural gas. The refrigerants used at each stage are generally selected as propane, ethylene (ethane), and methane. The advantages of this process include: low energy consumption; a single chemical as refrigerant for each stage; mature and stable technology. However, due to the use of three independent vapor compression refrigeration processes, the process is complex with many equipment units. Generally, the cascade liquefaction process is used in large liquefaction plants.

The mixed refrigerant cycle liquefaction process (MRC) is a process whose refrigerant is mixed multi-component refrigerant containing three to six components including C1

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