



Efficient biogas upgrading by a novel membrane-cryogenic hybrid process: Experiment and simulation study

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ABSTRACT

As a promising renewable fuel, biogas has been given widely attention. Upgrading raw biogas into high purity biomethane via a cost-effective route could facilitate the enhancement of heating value and combustion efficiency. In this study, biogas upgrading performance of a novel membrane-cryogenic hybrid system was investigated. The separation properties of two kinds of hollow fiber membranes (polyimide and polysulfone) were tested at different operating temperature, and then process simulation was conducted based on the experimental data. The experiment results indicated that operating membrane at the low temperature condition could be a potential method to improve biomethane purity (increased by 6.0% and 6.8% via polyimide and polysulfone, respectively), which was beneficial for combining membrane with cryogenic in the hybrid process. In addition, the simulation results showed that the total energy consumption of (polyimide) membrane-cryogenic hybrid process could be reduced to 0.8 MJ/kg-CH₄, when the cold energy was recuperated from liquefied biomethane (bio-LNG). It could be observed that the proposed novel membrane-cryogenic hybrid process could be a competitive alternative for the conventional biogas upgrading technologies to produce high quality (up to 98%) biomethane.

1. Introduction

Fossil fuel utilization generates multiple pollutants (e.g., particulate matter, sulfur dioxide, nitrogen dioxide, etc.) and greenhouse gas (e.g., CO₂), which can result in the issue of air pollution in the worldwide. In China, more than 70% energy is provided by fossil fuel (IEA [1]). Therefore, optimization of energy structure (such as developing renewable energy) is urgent and significant to mitigate pollution issues [2]. As a typical renewable energy, bioenergy might be a promising alternative to the conventional fossil fuel. In China, approximately 174.4–248.6 million dry metric tons of crop residues is discharged annually, which can provide sufficient feedstock for bioenergy exploitation [3].

As a typical bioenergy, biogas mainly generates from anaerobic digestion, and consisted of methane (CH₄), carbon dioxide (CO₂), small amount of water vapor and hydrogen sulfide [4]. It can be either used in combined heat and power engines (CHP) or as a natural gas substitute by removing CO₂ from CH₄. Until now, most biogase gas on-

farming CHPs, and the electrical power efficiency is less than 40% [5,6]. The high CO₂ content in raw biogas, not only reduces the calorific value and combustion efficiency, but also has an adverse influence on biomethane compression and pipeline transport [7]. In addition, since the pipeline specification of natural gas is CO₂ concentration below 2%, upgrading would be significant to produce high purity biomethane for efficient utilization [8].

Typical biogas upgrading technologies include water scrubbing, alkali solution absorption, pressure swing adsorption (PSA), membrane and cryogenic separation [9,10]. Biogas upgrading by alkanolamine solutions has been considered as one of the most mature technologies amongst the other available technologies [11]. The main advantages of amine-based absorption process are high capability to handle large amounts of feed stream, and easily to be retrofitted in the existing industrial plants without significant changes of installation [12–15]. However, the challenge of amine-based upgrading process is the high external energy input to regenerate rich solvents [16].

Recent year, membrane separation has been attracted increasing

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attention for CO₂ removal from different gas mixtures [17]. It has been expected to be an efficient technology to compete with chemical absorption due to their outstanding features, such as low operating cost, high operational flexibility and simple operating procedures [18–20]. According to the used materials, it can be divided into polymeric membrane, inorganic membrane and polymer-inorganic composite membrane. Among them, polymeric membranes are promising candidates for CO₂/CH₄ mixture separation due to their good chemical properties, thermal and mechanical stability [21–23]. However, it is difficult to purify the large amount of biogas by a standalone membrane module. Moreover, because of the low operating pressure, it requires more energy to achieve high CH₄ recovery with a high purity compared to water scrubbing process. Thus, a hybrid process via combining of membrane with other separation technologies (e.g., cryogenic) might be a promising method for biogas upgrading [24,25]. In previous works, membrane-cryogenic separation process has presented potential for efficient CO₂ recovery from flue gases [26–28]. By using membrane unit, bulk N₂ was removed at ambient temperature, and then the cryogenic unit could run more efficiently at a high CO₂ concentration (0.93 MJ_{electrical}/kg CO₂) [29]. However, it should be noted that there is few literatures related to biogas upgrading via membrane-cryogenic hybrid process, and the energy consumption of existing membrane-cryogenic hybrid processes should be further optimized to achieve the target 2 MJ_{thermal}/kg CO₂ (including both the capture and compression steps), which is often mentioned and corresponds to the European Union recommendations [29–31]. Meanwhile, recent works of Liu et al. [32,33] indicated that sub-ambient operating of membrane could facilitate the separation efficiency (the CO₂/N₂ selectivity increased up to 4 times than ambient), which provided a new option for membrane and cryogenic integration. Nevertheless, when mentioned cryogenic or cryogenic-based separation technologies, high energy consumption caused by cooling treatment is a crucial challenge. To avoid external energy utility, the cold energy can be obtained by integrating membrane-cryogenic biogas upgrading with liquefied methane (namely bio-LNG) regasification process, because upgraded biomethane is usually further liquefied to produce bio-LNG to facilitate transport and enhance energy density [34]. The substantial low temperature (about –160 °C) sensible heat and latent heat of vaporization makes it possible to reduce energy consumption of membrane-cryogenic hybrid biogas upgrading process (as shown in Fig. 1).

The objective of this study is to propose a novel cold membrane-cryogenic hybrid process for cost-effective biogas upgrading. For this purpose, the separation performance of two kinds of hollow fiber membranes (polyimide and polysulfone) at different low temperature condition was investigated. Based on the experimental results, process simulation was also conducted to evaluate the energy consumption and product properties of membrane-cryogenic hybrid process in large scale. To reduce the use of external energy, cold energy of bio-LNG can be recuperated by the hybrid process to replace the conventional refrigeration process, and thus, reducing the energy consumption. The detail cold energy utility and potential recovery strategies via heat integration are also discussed.

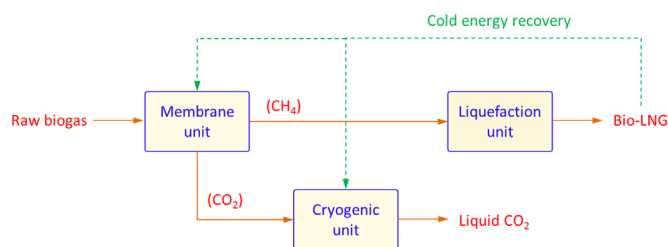


Fig. 1. Schematic of cold energy integration via bio-LNG in the novel membrane-cryogenic hybrid biogas upgrading process.

2. Experimental

2.1. Materials and methods

Polyimides have been shown to be the preferred class of polymer materials for gas separation membranes due to their excellent thermal and chemical properties as well as their superior separation efficiency compared to most other polymers [35]. Polysulfone has a preferential CO₂ adsorption ability compared to CH₄ due to higher critical temperature of CO₂ and the similarity of structure between sulfonyl group and CO₂ molecule [36]. In addition, polysulfone membrane also exhibits profitable properties such as good mechanical strength, compaction resistance and good thermal resistance [37]. Therefore, commercial polyimide (PI) (PA1010-P3, Permea China Ltd.) and polysulfone (PSF) hollow fiber membrane modules (NM-B02, Shanghai ACECOS technology CO., Ltd.) are tested in the experiments. The effective length and total effective area of polyimide hollow fibers are 30 cm and 2300 cm², respectively. In contrast, these parameters of polysulfone hollow fibers are 35 cm and 7200 cm². The mixed gas is composed of CH₄ (60%) and CO₂ (40%), which is used to simulate the main components of biogas. The pressure of the feed and retentate gas is 0.4 MPa and 0.1 MPa. The operating temperature is varied in the range of –30 to 40 °C to investigate its influence on the separation performance of membrane.

The permeability of the mixed gas was calculated by using the following equation:

$$J_i = \frac{Q_{y_i}}{A(P_{f_i} - P_{p_i})} \quad (1)$$

Where J_i is the membrane permeability, which can be expressed in GPU (1 GPU = 10^{–6} cm³ (STP)/cm² s cm Hg), Q is the gas permeation flow, y_i is the mole fraction of compound i in the permeation stream. A is the effective membrane area for permeation. P_{f_i} and P_{p_i} are the pressure of component i in feed gas and permeation gas, respectively.

The CO₂/CH₄ selectivity can be expressed as:

$$\alpha_{CO_2/CH_4} = \frac{J_{CO_2}}{J_{CH_4}} \quad (2)$$

Where J_{CO_2} and J_{CH_4} are the permeability of CO₂ and CH₄, respectively.

The CH₄ recovery can be calculated as :

$$R = \frac{Q_p \cdot y_{CH_4}}{Q_p \cdot x_{CH_4} + Q_r \cdot y_{CH_4}} \quad (3)$$

Where Q_p and Q_r are the flow rate of permeate and retentate gas, respectively. x_{CH_4} and y_{CH_4} are the CH₄ concentration of permeate and retentate gas, respectively.

2.2. Experimental setup and procedure

The schematic of the experimental setup for cold membrane performance evaluation is presented in Fig. 2. Temperature of the system is controlled by a low temperature incubator (LRH-100CB, Shanghai Yiheng Scientific Instrument Co., Ltd). K-type thermocouples are used to detect the temperature of ambient, permeate and retentate side, and the values are recorded in the temperature recorder (TR-W500, KEYENCE). CH₄/CO₂ binary gas is introduced into the system via tube feeding when the temperature decreased to the setting value. The pressure of feed gas, permeate and retentate streams are displayed on the pressure transducer (CYYZ13, Beijing Star Sensor Technology Co., Ltd). The flow rates of permeate and retentate streams are measured by rotor flow meter. The flow rate of the retentate side stream is controlled by the needle valve. Finally, the composition of permeate and retentate streams is analyzed by the Gas Chromatograph (GC).

The experimental temperature was initially kept at the pre-determined value (varied from –30 to 40 °C) via the low temperature

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