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Intensification of CO_2 separation performance via cryogenic and membrane hybrid process — Comparison of polyimide and polysulfone hollow fiber membrane

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ABSTRACT

Cryogenic-membrane hybrid CO_2 capture process has been considered as a promising alternative for the standalone processes to mitigate the critical challenge of energy penalty. In this work, two different commercial hollow fiber membranes were tested at low temperature condition to find the possibility of consisting a novel cold-membrane-cryogenic hybrid process. The gas permeation properties of membranes were evaluated at different operating temperature, CO_2 concentration and stage cut. The experimental results indicated that cryogenic condition could facilitate to improve the CO_2/N_2 selectivity with a minimum permeance reduction. Specifically, with the operating temperature dropped to −20 °C and feed pressure set at 0.7 MPa, the CO_2 selectivity of polyimide membrane increased by 166% with the permeance declined by 31%. Meanwhile, the CO_2 recovery and purity of polyimide membrane achieved 92% and 49% at the temperature of −20 °C and pressure 0.7 MPa, respectively. It can be observed that operating membrane at cold condition could be a promising approach to intensify the CO_2 separation, and thus decrease the energy consumption of subsequent CO_2 liquefaction or desublimation in the hybrid process.

1. Introduction

Greenhouse gases produced by human activity are mainly carbon dioxide, methane, ozone, nitrous oxide and chlorofluorocarbons etc. Among them, CO_2 accounts for 70% of total emissions, which leaded to the critical global climate issues, such as the greenhouse effect [1,2]. According to report of International Energy Agency [3], the average CO_2 concentration in atmosphere has increased to 404.21 ppm in 2016. In the next 50 years, the CO_2 concentration in the atmosphere will increase by 75 mg/L/year [4]. Therefore, greenhouse gas emission control is an urgent strategy to mitigate global warming and climate change. Carbon capture, utilization and storage (CCUS) is an efficient pathway to reduce the concentration of CO_2 in the atmosphere, which is to capture CO_2 from industrial sources, and then transfer it into high valuable chemicals or fuels, or store it in a special geological structure for a long time [5,6].

Membrane separation, as one of the dominant gas purification technologies, has the potential to be a cost-effective approach to capture CO_2 from flue gas [7]. Compared to absorption and adsorption processes, membrane separation could avoid the energy penalty of solvent or sorbent regeneration [8]. Meanwhile, the low operating cost and small equipment size are also beneficial to retrofitting with large emission sites (e.g. coal-fired power plants) [9,10]. The major challenges of the post-combustion membrane separation strategy are the large flue gas volume, low pressure and diluted CO_2 conditions, which correspond to a low driving force, and thus high energy consumption might be required to achieve CO_2 purity and recovery targets for transport and storage [11]. Meanwhile, although single membrane process can realize 90% CO_2 recovery, a multi-stage process is often necessary to keep a high level of CO_2 purity [12]. Therefore, more efforts should be paid on intensification and optimization of the membrane processes.

Recently, combination of two or more standalone CO_2 capture technologies, namely hybrid process, has attracted more and more attention due to the high capture efficiency and low energy requirement potential [13]. Among different hybrid options, membrane-cryogenic

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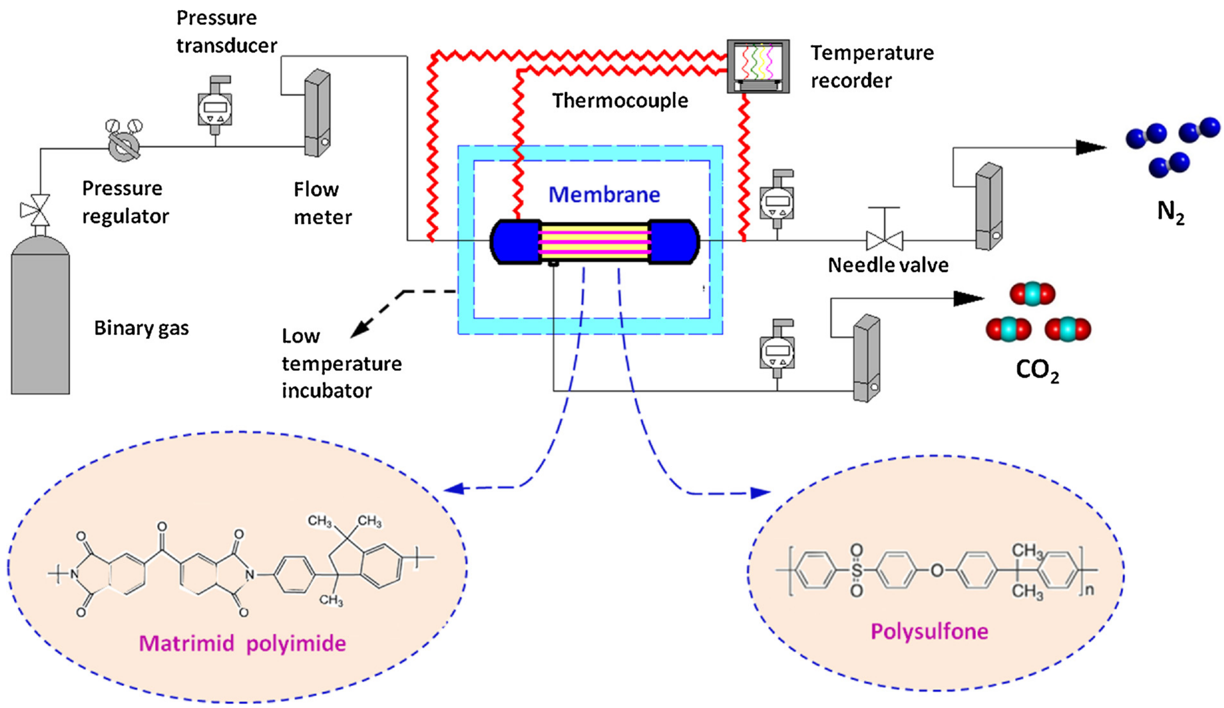

Fig. 1. Novel cryogenic-membrane hybrid CO_2 separation setup based on different hollow fibers.

arrangement is a promising alternative to the conventional CO_2 capture routes. It can increase the CO_2 concentration in the gas mixture by membrane unit, and then separate the enriched CO_2 in liquid or solid phase by a cryogenic unit [14]. Compared with MEA absorption and standalone membrane processes (>2.0 MJ/kg CO_2), the energy consumption of membrane-cryogenic hybrid route (lower than 3 $GJ_{thermal}$/ton CO_2 including compression of CO_2 to 110 bar) is competitive when CO_2 concentration in flue gases is in the range of 15%~30% [11,15]. This indicated that there may be a synergy between membrane and cryogenic units, and their combination could lead to a significant optimization with a minimal overall energy requirement. However, due to the sharp temperature drop from membrane and cryogenic units, the exergy destruction of hybrid process might be huge, and which would lead to a low energy efficiency and high energy consumption. Therefore, operating membrane at low temperature and followed by cryogenic CO_2 separation might be a novel arrangement of hybrid process. The potential advantage of this arrangement is that the temperature in cryogenic unit could be obviously increased due to more efficient preconcentration via cold-membrane unit, and thus total energy consumption for CO_2 capture could be obviously reduced. Meanwhile, up to now, most research of existing membrane-cryogenic hybrid processes is simulation work. There are few literatures related to the experiment validation of membrane-cryogenic hybrid processes [16].

In our previous work, a novel cold-membrane-cryogenic hybrid CO_2 separation system was designed with a low energy consumption (1.7 MJ/kg CO_2) [17]. In the novel hybrid process, membrane was operated at a low temperature condition to make full use of the excellent performance, and integrated with LNG (liquefied natural gas) regasification for cold energy reutilization, which was different from the conventional membrane-cryogenic hybrid technology. Low temperature could provide an unprecedented combination of CO_2 permeability and CO_2/N_2 selectivity [18–20]. The objective of this study was to explore the effect of low temperature condition on the performance separation of membrane. To further optimize separation performance of the proposed hybrid process, two different hollow fiber membrane modules (i.e. polyimide and polysulfone) are tested under cold condition. The effect of different operating variables (i.e. pressure, stage-cut, CO_2 concentration) on the cold membrane properties (i.e. permeability and CO_2 selectivity) and CO_2 separation performance (i.e. purity and recovery) of different hollow fiber modules is also investigated.

2. Experimental

2.1. Materials

The commercial membrane module with polyimide (PA1010-P3, Permea China Ltd.) and polysulfone fibers (NM-B02, shanghai ACECOS technology CO., Ltd.) are tested, respectively. A bundle of the hollow fibers are encased in the tubing, with both ends of the fiber bundle potted with epoxy resin to form a gastight tube sheet. The effective length of polyimide hollow fiber is 30 cm, which corresponds to a total permeation area of 2300 cm^2 in the membrane module. By contrast, the effective length and total permeation area of polysulfone hollow fiber are 36 cm and 7154 cm^2, respectively. The different compositions of CO_2 in the inlet feed stream, 5%, 10%, 15% and 20% CO_2 (balance by N_2), are used to simulate flue gas from different emissions.

2.2. Methods

2.2.1. Experimental procedure

The scheme of the hybrid separation setup is depicted in Fig. 1, and the detail configuration has been introduced in our previous work [17]. Two kind hollow fibers were individually placed in the low temperature chamber to test the gas permeability. Firstly, the operating pressure, stage-cut and CO_2 concentration in gas mixture were fixed to evaluate membrane properties and CO_2/N_2 separation performance of different membrane materials at low temperature. Then, the effects of different operating variables (pressure, stage-cut, CO_2 concentration) on the membrane properties and separation efficiency of the hybrid system were investigated. Pressure regulator was used to adjust feed pressure. The needle valve was used to control the retentate flow rate on the retentate side and obtain different stage-cut.

2.2.2. Parameter analysis

Stage cut is the measurement of the relative magnitude of permeability and feed flow rates, which is defined as: