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An experimental and modeling study of CO₂-selective membranes for IGCC syngas purification

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ABSTRACT

In recent years, integrated gasification combined cycle (IGCC) technology has gained significant attention for large-scale power generation with carbon capture. In an IGCC power plant, the syngas is typically directed to the gas turbine (for power generation) after CO₂ and H₂S removal. This work describes the transport properties of the CO₂-selective polymeric facilitated transport membranes for syngas purification at different pressures (2–15 bar) and 107 °C. These membranes contain amines as CO₂-carriers that are dispersed in a crosslinked polyvinylalcohol (PVA) matrix. Furthermore, a two-stage membrane process is proposed for CO₂ removal from syngas in an IGCC plant. A process modeling and cost-sensitivity analysis of the proposed two-stage membrane process was performed. The effects of feed pressure, temperature, membrane permeance, and selectivity on the overall increase in the cost of electricity (COE) were analyzed. An increase in the COE of 14–18% was estimated for 90% CO₂ removal and > 99% H₂ recovery using the two-stage membrane process in an IGCC power plant.

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1. Introduction

One third of the total CO₂ emissions come from the burning of coal as a fossil fuel [1]. Certainly, the CO₂ emissions from the coal-based power plants would play a dominant role in governing the global warming trends. Thus, there is an imminent need to restrict the CO₂ emissions from the coal-based power plants. There have been various studies focusing on comparing the integrated gasification combined cycle (IGCC) technology for power generation against the conventional pulverized coal (PC) and the oxy-fuel based combustion technologies. However, the development of carbon capture processes would govern the implementation of these technologies for the large-scale power generation [2,3].

In an IGCC power plant, coal or biomass is used to produce syngas (CO+H₂) by the gasification process. The syngas can be used as a feedstock for the production of value added chemicals or for the generation of electricity via the gas turbine or the fuel cell. Currently, there are some IGCC demonstration units but the technology has not been commercialized due to large capital cost for the gasification system and downstream syngas processing. However, it is economically more feasible to integrate a carbon

capture unit with an IGCC power plant relative to a conventional PC-based power plant. Therefore, IGCC power plants that are integrated with carbon capture are potential favorable alternatives for large-scale power generation.

Fig. 1 shows a process flow scheme of an IGCC power plant with carbon capture. Gasification is typically carried out at high pressures (> 50 bar) and at high temperatures (> 700 °C). Coal is gasified in presence of steam and oxygen to produce syngas. The oxygen is separated from air using the air separation unit (ASU) prior to the gasifier. The syngas from the gasifier is sent through a syngas quencher for tar and particulate removal prior to a series of water-gas-shift (WGS) reactors. The syngas is then passed through the WGS reactors where the carbon monoxide through reacting with steam is converted to carbon dioxide and hydrogen. The CO₂ and H₂S are required to be separated from the WGS reactor product stream for H₂ purification. The enriched H₂ stream is directed to the gas turbine for large-scale power generation. The H₂S separated from the syngas (using the Selexol[®] process) is directed to the Claus unit for the sulfur recovery, and the captured CO₂ is sent for sequestration purposes.

Membrane separation, solvent absorption and solid adsorption are three well-known processes for CO₂ removal [4]. Solvent absorption, at high pressures as well as low pressures, is well-developed among the three processes. The U.S. Department of Energy (DOE) have presented an independent techno-economic assessment (in their baseline document [5]) of the solvent absorption technologies for CO₂ removal in an IGCC, pulverized coal (PC)

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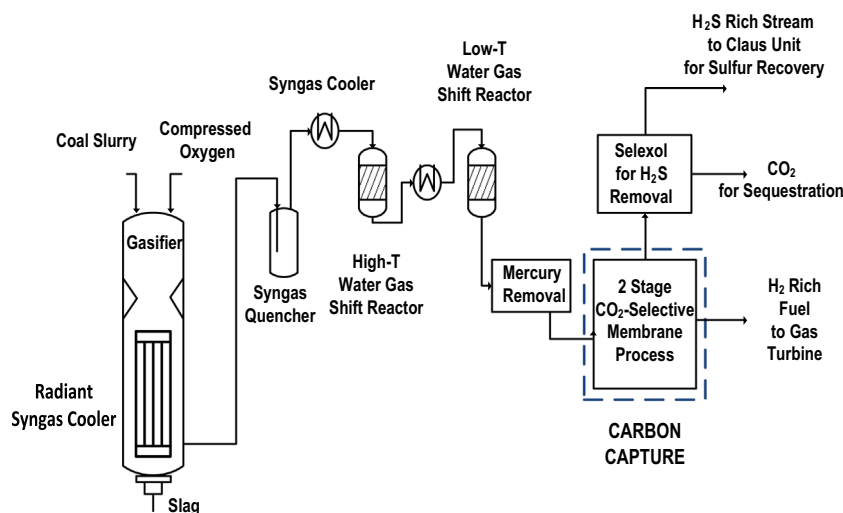


Fig. 1. Schematic representation of the IGCC power plant with carbon capture.

and natural gas combined cycle (NGCC) power plants. The state-of-the-art amine absorption process has been considered for CO₂ removal from the flue gas (at ~1 psig with ~14% CO₂ content, where 1 psi = 6894.76 Pa) in conventional PC power plants. Furthermore, a physical absorption based Selexol[®] process is considered for high-pressure CO₂ removal from syngas in an IGCC power plant. The presence of high pressure syngas and a higher CO₂ concentration (~35%) in the gas stream resulted in a lower cost to incorporate the CO₂ removal technology in an IGCC power plant as compared to the conventional PC power plant. The integration of state-of-the-art monoethanolamine process for CO₂ removal to a PC power plant resulted in approximately 86% increase in the cost of electricity (COE). On the other hand, the incorporation of the Selexol[®] process for carbon capture in an IGCC power plant resulted in about 39% increase in COE [5].

Absorption and adsorption processes are energy-intensive with significant parasitic power consumption. The membrane technology is a cost-effective process for H₂ purification due to lower energy consumption, lower capital cost, and operational simplicity. In an IGCC power plant with carbon capture, CO₂ needs to be separated from a high pressure gasification stream. A high pressure would mean a larger driving force for the membrane based CO₂ separation. However, separating CO₂ molecules selectively over the smaller-size H₂ molecules (2.89 Å; 1 Å = 1*10⁻¹⁰ m) at high pressures is a challenging task.

This work demonstrates attractive transport performances of CO₂-selective polymeric facilitated transport membranes for syngas purification. Furthermore, a process modeling and a systematic cost-sensitivity study of a two-stage membrane process for CO₂ capture from syngas in an IGCC power plant was performed. The IGCC power plant with the General Electric Energy (GEE) gasifier and no carbon capture (Case 1 of the baseline DOE document) was considered as the base case for further reference [5].

2. Membranes for CO₂ capture

A membrane process is considered in this work for separating CO₂ from syngas after the water-gas-shift reaction. The captured CO₂ is required to be recompressed for sequestration purposes. The enriched H₂ stream is sent to the gas turbine at high pressures (~32 bar or 3.2*10⁶ Pa) for power generation [5]. Since all the respective enriched gas streams (H₂-rich gas stream, captured CO₂ and H₂S streams) would be processed further at high pressures, it

would be more energetically favorable to operate the CO₂ and H₂S removal units at high pressures.

There are various CO₂- and H₂-selective membranes published in the literature for syngas purification at different pressures [6–10]. Inorganic palladium based membranes are reported for permeating H₂ at an almost infinite selectivity [4]. However, it suffers from severe sulfur poisoning (on exposure to H₂S) along with hydrogen embrittlement [11]. Moreover, it is difficult to scale up the fabrication of the state-of-the-art palladium membranes that could be rolled into large robust modules for practical applications. On the other hand, there are considerable developments in the field of polymer membranes for H₂ purification. Although the transport performances of polymeric membranes for H₂ purification are not as attractive, but it is commercially more viable to scale up the fabrication of polymer membranes as compared to the inorganic membranes.

There are two types of polymer membranes classified based on transport mechanisms: solution-diffusion and facilitated transport membranes. The CO₂-selective polymeric solution-diffusion membranes cannot be operated at high temperatures due to low CO₂/H₂ selectivity. However, the facilitated transport membranes appear to exhibit attractive transport performances at high operating temperatures [6–10]. The early facilitated transport membranes, supported liquid membranes and ionic liquids, showed attractive transport performances. But, they suffered from severe transport instabilities because of low resistances to the high pressure difference across the membrane, carrier leakage, solvent evaporation and slow reaction kinetics [4]. Recently, the water swollen polymeric facilitated transport membranes containing mobile and/or fixed-site CO₂-carriers have shown promising results at both the high-pressure and low-pressure conditions.

The schematic representation of the transport mechanism in a facilitated transport membrane is shown in Fig. 2. The reactive gas molecules (like CO₂) in the feed gas react reversibly with the carrier molecules in the polymer matrix. The gas-carrier reaction product diffuses from the feed-side to the permeate-side by the virtue of concentration gradient. The reactive gas molecules are released on the permeate side via the reversible reaction, resulting in the regeneration of the carrier molecules in the membrane.

There have been various amines in the form of mobile and fixed-site CO₂-carriers studied by Ho and co-workers at different pressures. Various amine carriers, namely potassium salt of α-aminoisobutyric acid (AIBA-K), glycine-Li, and lithium salt of N,N-dimethylglycine as mobile carriers, along with polyethylenimine (PEI) and commercial polyvinylamine (PVAm) as fixed-site

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