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## Optimization of membrane unit for removing carbon dioxide from natural gas

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#### **Abstract**

The aim of this paper is to find out the optimum configuration and design variables for the asymmetric membrane-based separation of carbon dioxide from natural gas for meeting the pipeline specification of 2% carbon dioxide. For this purpose, optimization of gas processing cost of the membrane unit having up to three stages is performed based on a fundamental model of the unit. It shows that there is no unique configuration that is always optimum irrespective of the values of carbon dioxide concentration and natural gas price. However, within certain ranges of the carbon dioxide concentration and the natural gas price, the optimum configuration may be unique and the minimum gas processing cost can be achieved by adjusting only the number of modules in each stage and the compressor power. In most cases, there is no significant cost difference between the two and three staged optimum configurations and the choice of a configuration may depend on engineering assessment. © 2006 Elsevier B.V. All rights reserved.

Keywords: Membrane unit optimization; Multi-stage membrane unit; Carbon dioxide removal; Natural gas treatment; Natural gas purification

#### 1. Introduction

Removal of carbon dioxide increases the calorific value and transportability of the natural gas. Carbon dioxide content in the natural gas obtained from gas or oil well can vary from 4 to 50%. On the other hand, purged gas from a gas-reinjected EOR (enhanced oil recovery) well can contain as much as 90% carbon dioxide. Before a natural gas rich in carbon dioxide can be transported, it must be pre-processed so as to meet the typical specification of 2-5% carbon dioxide. To meet such a specification, the natural gas is most commonly treated with an aqueous alkanolamine solution in absorption columns. The major advantages of the amine treatment are that it is a widely commercialized technology in which the hydrocarbon loss is almost negligible. However, the capital and operating cost shoots up very rapidly as the concentration of carbon dioxide in feed gas increases. An alternate technology for separation of carbon dioxide from the natural gas is based on the membrane.

Membrane consists of a polymer film that allows some molecules to permeate faster than other molecules. Molecules with higher diffusivity (or, lower size) and higher solubility in the film can permeate faster. Thus, carbon dioxide, hydrogen sulfide and water vapor can permeate faster than hydrocarbon molecules through cellulose acetate and many other similar polymeric membranes. To achieve high gas separation efficiency, the membrane is cast in an asymmetric form with very thin membrane film supported on a thick and porous polymeric layer on one side. Furthermore, membrane modules are fabricated in a very compact form as hollow fibers or spirally wound sheet. The residual feed stream from such a module is called the product or retentate, while the stream built up through permeation is called the permeate. For high-capacity applications, a membrane process unit can consist of multiple stages, each stage being constituted of one or more parallel modules and with internal recycling between stages. Apart from the modules, a membrane unit also requires pre-filter and pre-heater to prevent harmful deposit of dust, droplets and condensate on the membrane surface.

The membrane separation is normally associated with lower capital and operating costs than the amine treatment unit unless the area requirement is too high. With too large membrane area, a major concern is the loss of hydrocarbon to the permeate side via slow permeation. It is known that such a loss can be minimized by compressing the permeate and then recovering the hydrocarbons in a separate stage. However, this

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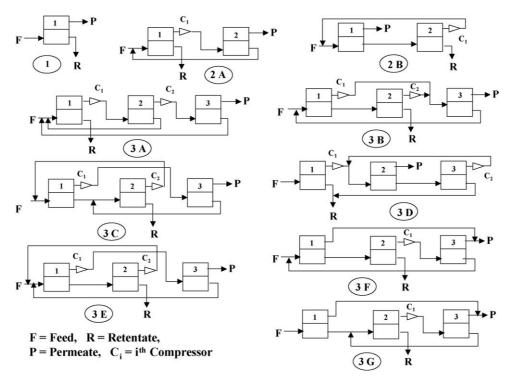


Fig. 1. Some probable optimum configurations of a membrane unit.

increases capital and operating costs of the membrane unit. Many studies have been performed in order to minimize these costs and make the membrane technology commercially viable. It has been suggested [1–3] that a feed gas with high carbon dioxide partial pressure (say, above 15 atm) can be treated in a hybrid unit. A pre-membrane unit will remove bulk of the carbon dioxide taking advantage of the high driving force for permeation. The product stream from the membrane unit can then be post-treated in an amine unit to meet the pipeline specifications. As described below, many reviews and studies have been reported on the optimal design of membrane-only unit for various gas separation applications including removal of carbon dioxide from the natural gas.

An excellent review on economics of applications of gas separation membranes has been provided by Spillman [3]. The author has shown the two-stage and three-stage configurations (e.g., configurations 2A and 3G in Fig. 1) that are more economical than the single stage configuration for carbon dioxide removal from natural gas. For applications in which the permeate stream is to be recompressed for further use (e.g., EOR application and hydrogen recovery from purge gas), the author has described how permeate can be generated at different pressures in a two-stage series configuration in order to minimize the permeate recompression cost. Configurations 2A and 3G was considered for treatment of natural gas containing 8–11% carbon dioxide [4] and EOR application for well-head gas containing 30–50% carbon dioxide [5], respectively.

Many authors have described rigorous optimization-based approach to select the optimal process configuration and other operating conditions for membrane units. Bhide and Stern [6] identified configuration 3G as the optimum choice for the feed

carbon dioxide concentration range 5-40% when natural gas price is 75 USD/K N m<sup>3</sup> (~2 USD/MMBTU) and asymmetric cellulose acetate membrane is used. Qi and Henson [7] also identified configuration 3G as the best choice for the feed carbon dioxide concentration range 5-95% and the natural gas price range 30-100 USD/K N m<sup>3</sup> when a membrane similar to asymmetric cellulose acetate is used. For natural gas price range 10–30 USD/K N m<sup>3</sup>, a different two-stage configuration (e.g., configuration 2B in Fig. 1) was found to be more economical. On the other hand, the authors identified another different threestage configuration (e.g., configuration 3E in Fig. 1) to be most economical choice for the feed carbon dioxide concentration range 0-50% when the permeate carbon dioxide concentration is restricted above 95%, in addition to the usual product concentration constraint of 2% carbon dioxide. Hao et al. [8] compared five different non-recycle configurations with two different types of asymmetric membranes for reducing carbon dioxide from 0–40% to 2% and hydrogen sulfide concentrations from 0–10% to 4 ppmv. Assuming the same natural gas price as in [6], they found the optimum configuration to be a CO<sub>2</sub>-selective membrane in the first stage in series with a H<sub>2</sub>S-selective membranes in the second stage.

In some gas processing applications, recovery of certain components through permeate stream is the most desired objective. For these applications, different innovative design configurations like CMC (continuous membrane column) and CRC (counter-current recycle cascade) have been proposed [9–10]. However, these configurations are not economical in natural gas treatment, where the processing objective is to produce natural gas with low concentrations of pollutants as the product stream [11]. Hence, it is beyond the scope of this paper to elaborate

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