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Structural optimization of membrane-based biogas upgrading processes

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ABSTRACT

Gas permeation membranes are well known for separating CO_2 and CH_4. Single stage processes cannot provide both, high product gas purity and high recovery at the same time, but multistage processes do. Commonly, the design of multistage gas permeation processes relies on heuristics and experience, so that often sub-optimal separation processes are designed. We apply a structural optimization approach to biogas upgrading processes. This systematic method determines the most profitable process layout including membrane areas and pressures required for two commercial membranes. Furthermore, we determine the optimal selectivity and CO_2 permeance following the upper bound of the Robeson plot. The process model is implemented using General Algebraic Modeling System (GAMS). For commercial membranes a three stage gas permeation process is the most profitable process which operates with only a single compressor. Optimizing the process layout together with the membrane properties a two stage process is the best process configuration. CO_2/CH_4 selectivities of approximately 120 are optimal. The impact of changed feed conditions and required product purity was investigated. The results highlight that for biogas upgrading commercial gas permeation membranes already provide powerful separation characteristics. The optimal membrane processes are capable to economically compete with conventional biogas upgrading techniques.

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1. Introduction

The depletion of fossil resources triggers the transition to renewable energies. Biogas upgrading will contribute to future energy supply. Contrary to solar and wind power, biogas is produced continuously. Biogas will considerably contribute to future energy supply as it is a renewable resource. Biogas upgrading refers to the separation of CH_4 and CO_2, where a CH_4 rich gas is polished so that it can be used as natural gas substitute [1]. Gas permeation membranes are well known for separating CO_2 and CH_4 [2–4]. Their application in biogas upgrading offers several advantages over conventional gas separation techniques. First the gas permeation operates with the upgraded product gas being at elevated pressure so that it can directly be injected into the natural gas grid [1]. Secondly gas permeation modules are robust and the process is simple, which is particularly suitable for on-farm application [5].

The objective of the study at hand is to design a membrane based biogas upgrading process based on structural process optimization. Here, (i) the optimal process configuration, (ii) the required membrane areas in the various stages and (iii) the pressure to drive the gas permeation process for a commercial membrane material are determined simultaneously. In a further step selectivity of the membrane material is optimized together with the process layout and the process conditions.

Process design often relies on heuristics and experience [6] which may result in suboptimal process configurations. The application of systematic methods for process design ensures the identification of the most profitable process configuration. Although superstructure optimization provides a systematic framework for the design of membrane based separation processes only limited work has been published. Sargent and Gaminibandara were the first who introduced the concept of structural optimization in designing distillation sequences. This concept has been adapted by El-Hawagi and Manousiouthakis who optimized mass exchange networks [7].

In the optimization of gas permeation processes Uppaluri et al. [8] studied the enriched oxygen production, hydrogen recovery from synthesis gas, and hydrogen recovery from refinery streams. They obtained new process designs which reduces the costs by 20% compared to process designs previously reported in the literature. They applied stochastic optimization using a simulated annealing algorithm.

Qi and Henson [9,10] report on CO_2/CH_4 separation in natural gas treatment and enhanced oil recovery. Here, only recycle compressors are investigated as the raw gas is already pressurized.

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They used GAMS along with the DICOPT++ algorithm to solve the MINLP. They even introduced discrete variables to account for a more realistic process design as membrane modules are available with fixed active membrane areas. They identified a four stage gas permeation process as the process configuration with the lowest gas treatment costs for natural gas polishing. For enhance oil recovery they also determined a four stage process as optimal process configuration as it has the lowest costs. In an additional publication [10] they extend the binary model to multicomponent model. The separation of CH_4, CO_2, H_2S and heavier hydrocarbons in natural gas treatment as well as the separation of CH_4, CO_2, H_2S, C_2H_6 and heavier hydrocarbons in enhanced oil recovery were investigated.

Kookos [11] does not only optimize the process layout and process conditions, but he also determines the best selectivity along with the best permeance for the production of nitrogen and oxygen from air. The upper bound correlation published by Robeson is applied to account for the membrane material performance. For the production of oxygen the comparison of an optimized membrane material and a commercial membrane material leads to the conclusion that impressive cost reductions are obtained.

We adapt the structural optimization approach to identify the most profitable membrane based biogas upgrading system. The process model is implemented in the General Algebraic Modeling System (GAMS). In a first step commercial gas permeation membranes are applied to the process optimization. The best process layout together with the optimal membrane areas and process conditions were calculated. In a second step the membrane's selectivity is an additional optimization parameter. The relation between selectivity and CO_2 permeance is determined according to the upper bound published by Robeson [12]. In contrast to Kookos [11] we applied the updated data for the upper bound correlation published in 2008. A three stage process is the optimal process configuration for the commercial membrane materials while a two stage process is the best layout for optimal membrane materials. An optimal CO_2/CH_4 selectivity in the order of 120 was most profitable for a process in which a single selectivity is applied to all membrane stages. When calculating the optimal selectivity for each membrane stage individually, a first highly selective stage is combined with highly permeable stage cleaning the permeate of the first stage.

The process model can easily be adapted to other gas separation problems such as helium production from natural gas, natural gas upgrading or air separation to determine the most profitable process configurations.

2. Process model

Commonly gas permeation processes include multiple membrane stages to achieve high gas purities and recoveries simultaneously. Favre [13] reported that more than three stages are usually not installed in industrial applications since these processes seem to be less efficient and cost demanding. In biogas upgrading it is rather unsuitable to use more than three stages as the complexity of the system increases rapidly and the application of such a system at a biogas site seems to be less robust. The process model implemented here is not limited to any number of membrane stages, but for the optimization of the biogas upgrading process a maximum number of three membrane stages is defined.

The gas permeation modules considered in the simulations are equipped with polymeric membrane materials to separate CO_2 and CH_4. CO_2 will permeate faster through the membrane so that the permeate is enriched in CO_2. Thus, CH_4 is enriched on the retentate side of the membrane which is particularly advantageous in biogas upgrading as the product gas is pressurized and can thereby directly be injected into the natural gas grid. Table 1 lists the raw gas conditions and the product gas requirements. A membrane CO_2/CH_4 selectivity of 60 is assumed based on a current patent [14] for a polyimide membrane material.

However, no information is given on the permeances of such a membrane material and we assume a CO_2 permeance of 60 GPU. In addition, a membrane material with selectivities reflecting the characteristics of a cellulose acetate membrane is investigated. The permeances are also listed in Table 1.

In general, feed gas conditions, product gas requirements and economic parameters such product gas pressure and energy costs considerably determine the optimal process configuration. Consequently, the optimal process configuration presented here, is identified by applying the boundary conditions set here. This process configuration is most likely not optimal for the entire range of parameters to be applied in biogas upgrading and boundary conditions have to be reviewed carefully. The objective of the optimization is to maximize the profit of the biogas upgrading plant. Four different types of unit operations are implemented and could be used in the process model. These are a gas permeation module to perform the separation and equipment to generate the driving force of permeation such as compressors, vacuum pumps, and pressure control valves. Fig. 1 shows the respectiveunit operations and illustrates the potential connections of the individual unit operation with other unit operations. Here only selected connections are presented. For the gas permeation model it is also possible to use a sweep gas on the permeate

Table 1
Feed and product gas conditions of a common biogas upgrading process. The permeances of the polymeric membrane materials are also listed. PI – polyimide; CA – cellulose acetate; 1 GPU = 2.7×10^{-3} m_N^3/m^2h bar.

	Unit	Value
Raw gas mole fraction CH_4	–	0.6
Raw gas mole fraction CO_2	–	0.4
Raw gas flow rate	$\frac{m^3(STP)}{h}$	150
Raw gas temperature	°C	20
Raw gas pressure	bar	1
Product pressure	bar	16
Product mole fraction CH_4	–	0.96
CH_4 permeance PI	GPU	1
CO_2 permeance PI	GPU	60
CH_4 permeance CA	GPU	3
CO_2 permeance CA	GPU	60

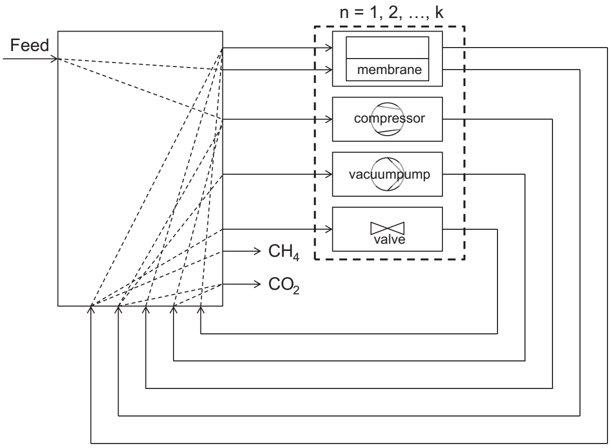

Fig. 1. Superstructure approach to optimize a membrane based biogas upgrading plant. Note that not all possible connections between the various unit operations are presented.

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