Renewable Energy 131 (2019) 1117–1127

Contents lists available at ScienceDirect

Renewable Energy

journal homepage: www.elsevier.com/locate/renene

Optimizing purity and recovery of biogas methane enrichment process in a closed landfill

S.Z.A. Seman, I. Idris, A. Abdullah, I.K. Shamsudin, M.R. Othman*

School of Chemical Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 20 December 2017
Received in revised form
14 June 2018
Accepted 15 August 2018
Available online 18 August 2018

Keywords:
Purity-recovery
Optimization
Membrane
Biogas methane
Carbon neutrality

ABSTRACT

Biogas calorific value upgrading into biomethane or methane-rich gas is an attractive choice for renewable energy alternatives. Exploitation of methane-rich gas from landfill has gained increasing traction recently due to its fuel source diversification potential towards carbon neutrality and energy security. In this paper, methane enrichment process in a membrane plant from a biogas mixture containing methane, carbon dioxide, nitrogen, oxygen, hydrogen sulfide and water vapor was modeled and compared with the actual plant data. Results from this work reveal an interesting relationship between purity-recovery and the process operating conditions. The model was capable of simulating the methane enrichment effectively with merely 0.07% error in predicting methane purity and 0.52% error in predicting methane recovery. The model allows optimization of methane purity from 87.26% to 99.87% and methane recovery from 91.63 to 99.49% at permeate to feed ratio (stage cut) of 0.30, without the need for re-staging of the plant.

1. Introduction

Biogas energy is increasingly gaining public and private sector attention lately due to its renewability and amid the global environmental concern related to the anthropogenic greenhouse gases emission from fossil fuels. Recently, a company which is involved in solar energy decided to switch its focus to biogas energy. This is due to the more sound financial strategy of the biogas energy than the financial investment of the solar energy that does not make sense [1].

Biogas is a product resulting from anaerobic decomposition of organic matter and comprises mainly of methane (CH_4), carbon dioxide (CO_2), smaller traces of acidic gases and impurities such as hydrogen sulfide (H_2S), nitrogen (N_2), water vapor (H_2O) and traces of other volatile organic compounds (VOC) [2]. The concentration of CH_4 in the biogas depends largely on the source of organic substrate and the decay conditions. CH_4 composition from poultry waste, for an example, can vary from as low as 40% to higher than 70% [3,4]. A biogas containing 70% CH_4 and 30% CO_2 by volume was produced from fermentation of fatty acid and used in methane enrichment process utilizing hollow fiber membrane [5]. In another study [6], biogas containing 55.00% CH_4, 38.90% CO_2, 0.40% O_2, 5.00% N_2, 0.002% H_2S and 0.66% H_2O (in mole percent) was produced from naturally decomposed organic waste in a closed landfill at Sebenza, in the Ekurhuleni Municipality of South Africa. The gas was pretreated prior to use in methane enrichment process using a single and dual stage membrane configuration.

Methane enrichment from biogas is important in order to enhance the calorific value of the gas, to achieve low emission requirement of nitrogen oxide and other gases upon combustion and to meet the fuel quality for injection to the natural gas grid. Pure methane has a calorific value of 9100 kcal/m^3 at 15.5 °C and 1 atm. The calorific value of biogas varies from 4800 to 6900 kcal/m^3. In order to achieve the standard calorific value of 5500 kcal/m^3, the treatment techniques like conventional amine or solvent absorption system or membrane separation are required [7].

There is a concern surrounding the feasibility of the current methods of methane enrichment process. The current absorption technique, conventionally used in natural gas sweetening to strip off the unwanted impurities employs expensive and environmentally unfriendly solvents, has not reached the acceptable efficiency level that is important for commercial applications of the biogas [8]. Variety of methods have been reviewed and proposed to enrich and purify methane from raw biogas mixture containing carbon dioxide that not only meet the transportation fuel quality and grid injection standards for power generation but also reach sufficient level of

* Corresponding author.
E-mail address: chroslee@usm.my (M.R. Othman).

https://doi.org/10.1016/j.renene.2018.08.057
0960-1481/

efficiency for sound financial investment strategy. Some of these methods include the upgraded solvent absorption system [10], pressure swing adsorption [11–15], cryogenic separation [16,17] and membrane technology [5,6,10–13,18–31]. Another review also addresses concern on low economic and energy efficiency of the current biogas upgrading and treatment techniques [9].

Increasing interest in biogas upgrading has opened up new frontiers in the development of membrane technology for gas separation. Membrane separation technology has been identified as an attractive choice due to its simplicity, favorable economics and low carbon footprint. It has been increasingly researched in numerous environmental [12,16,22,26] and energy processes from power plant such as CO_2 capture [11,13–16,18], VOC recovery [16,17,21] and biogas upgrading [2,5–10,18]. There are different types of membranes being used for gas separation. The most commonly researched membranes for separation of methane from carbon dioxide and other gases include dense and microporous polymers [5,6], mixed matrix membranes [18], molecular sieve membranes [28,29] and mesoporous and microporous inorganic membranes [20–24].

The efficiency of the membrane system depends largely on the recovery and purity of the methane gas. Higher than 90% purity and 90% recovery are needed in order to achieve the economics of operation. A hollow fiber membrane with CO_2/CH_4 selectivity of 246 reportedly [32] achieved 99.5% recovery and 97.5% purity. This achievement was observed for biogas feed mixture containing 30–50% CO_2 at feed pressure of 10 bars and feed flow rate of 300 Nm^3/h. Considering methane price of 0.78 $/$Nm^3$ and membrane lifetime of 15 years, the techno-economic analysis showed that the payback time was 1.6 months [32], despite the pressure is considered unnecessarily high.

There is always a trade-off between purity and recovery. Higher purity is achieved at the expense of lower recovery. The level of both purity and recovery could be improved by increasing the pressure, but increasing the pressure means more compression load is required and this would translate into higher operating cost. In balancing the need to achieve the highest purity and recovery at the lowest possible energy consumption, process optimization plays a significant role. Optimization is regarded as the most advanced stage in technological maturity level. It is important in maximizing production while conserving the environment. Optimization is the essence of the latest industrial revolution (IR-4) in many modern industrial processes. Here, optimization can be realized by understanding the process from the perspective of a mathematical model and simulation.

There is an attempt to simulate purity and recovery of methane and carbon dioxide from an actual plant operation in the past. During the methane upgrading process in the previous work [6], a hollow fiber membrane was employed to achieve the desired fuel grade biomethane. The feed and permeate pressures applied to the membrane were 9.90 bars and 1.08 bars, respectively at feed flow rate of 80 m^3/h. The process was simulated using ChemCAD, a steady state process simulator from Chemstation. The simulation process encompassed single stage without recycle (SSWR) and dual stage with permeate recycle (DSPR). The mathematical model used to predict the results, however, gave considerably large error for SSWR. Correction was made to reduce the error by restaging the entire process using DSPR. This strategy, however, is not recommended since it misrepresents the actual membrane plant setting. In the actual plant, DPSR was not present.

In this study, a complete mixing was proposed to model the methane upgrading process. In a complete mixing model, the material balance of each gas species in the mixture was maintained and the permeability of all the component species was constant throughout the membrane. The model was applied to find the relationship between purity-recovery and the membrane operating conditions. The purity-recovery of CH_4 in reject and the purity-recovery of CO_2 in permeate were optimized using a Response Surface Methodology (RSM). This effort should be able to fill the gap in the literature for better understanding of the membrane separation in biomethane enrichment/upgrading process.

2. Materials and methods

A complete mixing model assumes that there is negligible pressure drop in the feed and permeate sides and that the permeability of all the component gas is constant throughout the membrane length. The permeability and mole fraction of each gas species are given in Table 1. The permeability values are obtained from the previous work [6]. The membrane used was a fluorinated polyimide (6FDA-TMPA) hollow fiber. The schematic diagram of the hollow fiber membrane for biomethane enrichment in the reject stream is given in Fig. 1 [6].

Table 2 shows CH_4 and CO_2 purity and recovery using single stage without recycle (SSWR), dual stage with permeate recycle (DSPR) obtained from the previous attempt [6]. The actual purity and recovery of CH_4 in the reject measured at plant using single stage membrane module are 87.20% and 91.16%, respectively. Whereas, the purity and recovery of CO_2 in the permeate stream measured at plant are 54.00% and 39.87%, respectively.

The overall material balance and permeate through the membrane following the complete mixing model are given by Ref. [33],

$$q_f = q_p + q_o \tag{1}$$

$$q_p = \frac{KA}{t}(P_h - P_l) \tag{2}$$

where, q_f is the total feed flowrate, q_p is permeate flowrate, q_o is reject flowrate, K is permeability of the gas species, A is membrane area, t is membrane thickness, P_h is feed pressure and P_l is permeate pressure. The membrane properties and process parameters obtained previously from a plant [6] are given in Table 3.

Rate of diffusion based on the permeability of each gas species is given by Ref. [33],

$$q_p \bullet x_{pi} = K_i \frac{A}{L}(P_h \bullet x_{oi} - P_l \bullet x_{pi}) \tag{3}$$

$$q_p \bullet x_{pj} = K_j \frac{A}{L}(P_h \bullet x_{oj} - P_l \bullet x_{pj}) \tag{4}$$

$$q_p \bullet x_{pk} = K_k \frac{A}{L}\left(P_h \bullet x_{ok} - P_l \bullet x_{pk}\right) \tag{5}$$

$$q_p \bullet x_{pl} = K_l \frac{A}{L}\left(P_h \bullet x_{ol} - P_l \bullet x_{pl}\right) \tag{6}$$

Table 1
Properties of components in the biogas mixture and permeability of the gas across Polyimide TMPA-6FDA membrane [6].

Gas	Molecular Weight (g/mole)	Mole fraction [6]	Permeability [6] (Barrer)
CH_4	16.04	0.55000	28.2
CO_2	44.01	0.38934	440.0
N_2	28.02	0.05000	35.6
O_2	32.00	0.00400	111.0
H_2S	34.08	0.00002	1.0
H_2O	18.02	0.00664	1.0