Applied Energy xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Applied Energy

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

Techno-economic evaluation of biogas upgrading using ionic liquids in comparison with industrially used technology in Scandinavian anaerobic digestion plants

Yujiao Xie [a,b,*], Johanna Björkmalm [c], Chunyan Ma [a], Karin Willquist [c], Johan Yngvesson [c], Ola Wallberg [d], Xiaoyan Ji [a,*]

[a] Energy Engineering, Division of Energy Science, Luleå University of Technology, 971 87 Luleå, Sweden
[b] School of Chemical Engineering, Shandong University of Technology, Zibo 255049, China
[c] SP Technical Research Institute of Sweden, Box 857, 501 15 Borås, Sweden
[d] Department of Chemical Engineering, Lund University, Box 124, 221 00 Lund, Sweden

HIGHLIGHTS

- The process performances with four absorbents for biogas upgrading were compared.
- The economic evaluation for biogas upgrading using ILs was performed.
- The industrial data from biogas upgrading plants was collected.
- Simulation results of IL technique were compared with data from commercial facilities.

ARTICLE INFO

Article history:
Received 6 January 2017
Received in revised form 30 June 2017
Accepted 17 July 2017
Available online xxxx

Keywords:
Techno-economic evaluation
Process simulation
Biogas upgrading
Ionic liquids
Anaerobic digestion

ABSTRACT

The process of biogas upgrading with ionic liquids, i.e. pure 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([bmim][Tf_2N]), aqueous choline chloride/urea (ChCl/Urea), and aqueous 1-allyl-3-methyl imidazole formate ([Amim][HCOO]), was simulated in Aspen Plus and compared with the conventional water scrubbing upgrading technique. The comparisons of the performances on the amount of recirculated solvents and energy usage show the following order: aqueous [Amim][HCOO] < aqueous ChCl/Urea < [bmim][Tf_2N] < water. Six different co-digestion plants (anaerobic digestion, AD, plants) were surveyed to acquire data for comparison. The selected plants had different raw biogas production capacities and produced gas with differing methane content. The data confirmed the simulation results that the type of substrate and the configuration of AD process are two factors affecting energy usage, investment cost, as well as operation and maintenance costs for the subsequent biogas upgrading. In addition, the simulation indicated that the energy usage of the ionic liquid-based upgrading was lower than that of the conventional upgrading techniques in Scandinavian AD plants. The estimated cost including investment, operation and maintenance for the ionic liquid technology showed to be lower than that for the water scrubbing upgrading process.

1. Introduction

Energy security and climate change concerns have resulted in increased attention towards renewable energy. Biogas is a promising renewable energy alternative due to the flexibility of anaerobic digestion (AD) where a spectrum of organic substrates (such as manure, sewage sludge, energy crops, the organic fractions of household and industrial waste) can be used [1,2]. In Sweden, it has been estimated that the greenhouse gas (GHG) savings can be up to 80–90% if biogas is used as vehicle fuel or for replacing natural gas by injection into the gas grid [3]. Consequentially, the biogas production in the European Union has steadily increased over the past years [4].

Biogas consists mainly of methane and carbon dioxide, with small amounts of hydrogen sulfide, water, hydrogen, nitrogen and oxygen. The methane content in AD-produced biogas ranges

---

[*] Corresponding authors at: Energy Engineering, Division of Energy Science, Luleå University of Technology, 971 87 Luleå, Sweden (Y. Xie and X. Ji).
E-mail addresses: yujiaoxie@sdut.edu.cn (Y. Xie), xiaoyan.ji@ltu.se (X. Ji).

http://dx.doi.org/10.1016/j.apenergy.2017.07.067
0306-2619/© 2017 Elsevier Ltd. All rights reserved.

Please cite this article in press as: Xie Y et al. Techno-economic evaluation of biogas upgrading using ionic liquids in comparison with industrially used technology in Scandinavian anaerobic digestion plants. Appl Energy (2017), http://dx.doi.org/10.1016/j.apenergy.2017.07.067

between 50 and 70% [4] and consequentially need to be upgraded to at least 97%, according to the Swedish regulations, SS 155438:2015 [5], in order to be used as vehicle fuel or be injected to the gas-grid. Similarly, there is ongoing standardization work within the EU regarding biomethane for transport applications and injection to natural gas pipelines [4].

To upgrade biogas, CO_2 separation from raw biogas is a main step. Different technologies have been developed, such as water scrubbing, membrane separation and chemical absorption [6–11]. High pressure water scrubbing is used to separate CO_2 based on physical absorption. This technology has been developed, implemented and commercialized in several European countries [12]. The main deficiency of this technology is the low absorption capacity of CO_2, resulting in large amount of water [13]. In membrane separation, the multi-stage membranes are always necessary in order to achieve high degree of separation, which increases the manufacture and capital cost [14]. In the chemical absorption, the amine-based solvents are widely used, while the amine technology is energy-intensive with the deficiencies of volatility, degradation, and corrosion of amine [15]. Due to the drawbacks of these technologies, it is necessary to explore new technologies for biogas upgrading.

Ionic liquids (ILs) have received much attention for CO_2 separation since Blanchard et al. [16] reported the high CO_2 solubility in IL in 1999. ILs have shown a great potential to be used as a solvent for biogas upgrading due to the high CO_2 solubility/selectivity, low energy requirement for solvent regeneration and other unique properties (e.g. negligible vapour pressure). Bidart et al. [17] investigated biogas upgrading using common and functionalized ILs and concluded that the advantage of ILs is the low volatility and chemical stability, which facilitate biogas upgrading. Zhang et al. [18] studied the thermodynamic and mass-transfer properties of ILs and recommended 50 wt% [bmim][NO_3] + 50 wt% polyethylene glycol dimethyl ethers mixture as solvents for biogas upgrading. Suggestions on developing new ILs to further enhance selectivity and absorption capacity were provided. Xu et al. [13] found that the energy usage of conventional IL technology for biogas upgrading is similar to that for water scrubbing but much lower than that for aqueous monoethanolamine (MEA) scrubbing.

In our previous work [19], the conceptual processes for biogas upgrading using three imidazolium-based ILs were simulated, and the simulation shows that the energy usage for [bmim][Tf_2N] scrubbing is the lowest. Meanwhile, novel ILs have been studied in our research group. 1-allyl-3-methyl imidazole formate ([Amim][HCOO]) was synthesized as a tailored sorbent for CO_2 separation [20], and aqueous choline chloride/urea (ChCl/Urea) was studied as a novel eutectic IL-based solvent to separate CO_2 because of low price and environmentally benign [21]. However, how these solvents compare to each other and to conventional upgrading technologies for biogas upgrading in respect to energy efficiency and cost have not yet been studied.

The operation of biogas upgrading depends on the composition and flow rate of raw biogas. Under industrial conditions, the composition and gas flow rates vary significantly between plants [4]. To assess the performance of upgrading process, it is preferable to have industrial data as the input of the simulation and as benchmarking data to compare the simulation results. The type of data that is needed and how to calculate key parameters (e.g. energy usage, production cost) are important parameters for this comparison. However, comparison of simulation results and data from commercial biogas upgrading facilities are to our knowledge lacking in the literature.

In this work, the properties of four absorbents (water, [bmim][Tf_2N], aqueous ChCl/Urea, and aqueous [Amim][HCOO]) were compared, and the process simulations on biogas upgrading with these four absorbents were performed. The energy usage of aqueous [Amim][HCOO] scrubbing was further investigated via sensitivity analysis with the input from Swedish and Norwegian industrial biogas processes and an economic evaluation was conducted. The results were compared to industrial data from different biogas upgrading processes.

2. Methodology

2.1. Absorbents and properties

The properties of four absorbents mentioned above were compared before conducting process simulation. Based on our preliminary investigation [22], it is found that [bmim][Tf_2N] is the most promising IL with the lowest energy demand among the investigated conventional imidazolium-based ILs. The investigation shows that the aqueous ChCl/Urea with 50 wt% ChCl/Urea is the recommended solvent for biogas upgrading [21]. The aqueous [Amim][HCOO] with 90 wt% [Amim][HCOO] was chosen for the further study considering the viscosity [20]. Therefore, H_2O, (50% ChCl/Urea + H_2O), (90%[Amim][HCOO] + H_2O) and [bmim][Tf_2N] were chosen as absorbents to be studied in this work.

In our previous work [19–21], the database of thermophysical properties for [bmim][Tf_2N] [19], dry/aqueous ChCl/Urea [21], and dry/aqueous [Amim][HCOO] [20] was implemented in Aspen Plus based on the consistent experimental data. The binary interaction parameters of Non-Random Two-Liquid (NRTL) model describing the properties in the liquid phase for CO_2-IL, CH_4-IL and IL-H_2O and the parameters describing the Henry's constant in IL for CO_2 and CH_4 were obtained from the fitting of the experimental data and implemented into Aspen Plus. The details of these parameters are listed in the supporting information. The comparison of these predicted results using Aspen Plus with the available experimental data had also been conducted in our previous work [19–21]. It showed that the predicted values are reliable.

In this work, the properties including density, viscosity and CO_2 loading of four absorbents were obtained by using the property analysis model in Aspen Plus and compared with each other. For the gas selectively, the flash mode based on NRTL-RK (Non-Random Two-Liquid - Redlich–Kwong) model was used to calculate the mole fractions of CH_4 and CO_2 in the liquid phase (i.e. x_{CH4}, x_{CO2}) with the parameters of RK (Redlich–Kwong) taken from Aspen databank. The expression of selectivity is shown in Eq. (1).

$$s = \left(x_{CO_2}/x_{CH_4}\right)_{T,P} \tag{1}$$

2.2. Process description

AD is an effective method to produce CH_4 from organic substrates [23,24]. The substrate used for digestion is collected and pretreated in order to increase the process efficiency. The pretreated substrate is entered into the fermentation tank to generate raw biogas. The composition of raw biogas, with respect to CH_4 concentration, and the gas flow rate are dependent on the type and amount of substrate and the configuration of the anaerobic digestion process, e.g. the retention time in the digester. The raw biogas is further upgraded in order to increase the energy content by removing CO_2 and other impurities. The upgraded biogas with >97% CH_4 is termed as biomethane, and it can be used as vehicle fuel or further converted to industrial chemicals.

The conceptual process to upgrade biogas with ILs has been described in our previous work [19,21]. As shown in Fig. 1, it consists of the absorber for CO_2 absorption, flash tank to decrease methane loss and the desorber for solvent regeneration tank. The raw biogas is compressed and injected into the bottom of the absorber, and the liquid solvent is sprayed from the top of the

Please cite this article in press as: Xie Y et al. Techno-economic evaluation of biogas upgrading using ionic liquids in comparison with industrially used technology in Scandinavian anaerobic digestion plants. Appl Energy (2017), http://dx.doi.org/10.1016/j.apenergy.2017.07.067