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Techno-economic analysis and performance comparison of aqueous deep eutectic solvent and other physical absorbents for biogas upgrading

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HIGHLIGHTS

- The performances of one novel and 3 conventional absorbents were compared.
- The economic evaluation of 4 solvents for biogas upgrading were performed.
- Both equilibrium and rate-based approaches in Aspen Plus were used for comparison.
- The economics in either plant retrofit or building-up new process were studied.

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ABSTRACT

Biogas has been considered as an alternative renewable energy, and CO_2 removal from raw biogas (i.e. biogas upgrading) is needed for producing biomethane to be used as vehicle fuels or injected into the natural gas grid. Biogas upgrading with physical absorbents, such as water and other commercial organic solvents, is simple, efficient and with low energy requirements for regeneration. Recently, deep eutectic solvents (DESs) with nonvolatility, nonflammability and low price have been reported as promising alternatives to replace conventional physical absorbents in many research areas including biogas upgrading. However, the performances of these physical solvents including conventional physical absorbents and DES-based solvents have not been evaluated and compared with each other. In this work, the properties of 4 physical solvents (i.e. water, dimethyl ether of polyethylene glycol (DEPG), propylene carbonate (PC), and aqueous DES (AQ_{DES})) were compared. Furthermore, a conceptual process was developed to upgrade biogas with these solvents and simulated based on Aspen Plus in order to conduct performance comparison. The simulation results of energy utilization, the amount of recirculated solvents and the diameters of absorber and desorber were analyzed and compared based on equilibrium and rate-based approaches, respectively. The simulation results based on the rate-based approach were further used to estimate the costs of biogas upgrading process with a same raw biogas capacity for comparison. Meanwhile, the specific cost of biogas upgrading process with a same size of equipment was also evaluated. The results show that the CO_2 solubility, selectivity and viscosity are three more important properties, providing valuable information for developing novel physical solvents for CO_2 separation. The simulation results show that the equilibrium and rate-based approaches result in different conclusions, especially when the solvent viscosity is relatively high, and the rate-based approach is preferable. Based on the simulation results from the rate-based approach, the performances of AQ_{DES} and PC are similar with a same amount of energy utilization, that is around 11% lower than water, and DEPG is inferior to water. For the case with the same gas capacity, the total annual costs of biogas upgrading process with these solvents show the following order: DEPG > $AQ_{60wt.\%DES}$ > water > $AQ_{50wt.\%DES}$ ≈ PC. For the case with the same size of equipment, compared to water, the total specific costs of biogas upgrading process with PC and $AQ_{50wt.\%DES}$ decrease by about 30% and 45%, respectively, and the treated biogas capacities increase to 1.5 and 2 times, respectively. In general, both PC and $AQ_{50wt.\%DES}$ show better performance than the other solvents. Considering that DES is an environmentally benign solvent, and the performance of DES can be greatly improved by further designing, it is more promising.

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Nomenclature

Abbreviations

DESs	deep eutectic solvents
ILs	ionic liquids
DEPG	dimethyl ether of polyethylene glycol
PC	propylene carbonate
AQ_{DES}	aqueous deep eutectic solvent
$AQ_{60wt.\%DES}$	aqueous deep eutectic solvent with 60 wt.% of DES
$AQ_{50wt.\%DES}$	aqueous deep eutectic solvent with 50 wt.% of DES
HPWS	high pressure water scrubbing
PSA	pressure swing adsorption
CA	chemical absorption
OPS	organic physical scrubbing
MB	membrane separation
CS	cryogenic separation
TAC	total annual cost
ACC	annual capital cost
O&MC	operation and maintenance cost
ACC	annual capital cost
TCC	total capital cost
EC	equipment cost
TDC	total direct cost
IC	indirect cost
FCI	fixed capital invests
TIC	total indirect cost
OL	operating labour
TFC	total fixed charge
TDPC	total direct production cost
TGE	total general expenses
R&D	research and development
PEC	bare purchased equipment cost
HETP	height equivalent of a theoretical plate
S	size characteristic values of the equipment
Mw	molecular weight, $g{\cdot}mol^{-1}$
H_{CO2}	Henry's constant of CO_2
H_{CH4}	Henry's constant of CH_4
L/G	the ratio of the volumetric flow rate of the absorbent to that of raw biogas, $m^3{\cdot}m^{-3}$
PG	product gas
$\Delta P/z$	pressure drop per height, $N{\cdot}m^{-3}$
$E_{CH4\ loss}$	the energy utilization related to the CH_4 loss
$Nm^3{\cdot}h^{-1}$	normal meter cubed, gas volumetric flow rate at normal conditions of 273.15 K and standard atmosphere

1. Introduction

Biogas has been considered as an alternative renewable energy, and CO_2 removal from raw biogas (i.e. biogas upgrading) is needed for producing biomethane that can be used as vehicle fuels or injected into the natural gas grid [1–4]. A number of technologies have been developed and commercialized to upgrade biogas, for example, high pressure water scrubbing (HPWS), pressure swing adsorption (PSA), chemical absorption (CA), organic physical scrubbing (OPS), membrane separation (MB), cryogenic separation (CS), and so on [5,6]. It has been reported that physical solvents tend to be favored over chemical solvents when the concentration of acid gas is high [7]. In addition, physical solvents are non-corrosive and thus require only carbon steel construction.

In a physical absorption process such as HPWS or OPS, a solvent is used to absorb CO_2 from biogas with a small loss of CH_4 on the basis of physical effect. HPWS is the most widely used biogas upgrading technology in European countries [8,9]. For OPS, dimethyl ether of polyethylene glycol (DEPG) is a typical organic physical absorbent, which is known as Selexol and Genosorb [10]. Compared with water, the high solubility of DEPG can decrease the amount of absorbent, while the low selectivity of DEPG usually causes high CH_4 loss and then the additional thermal energy is required to increase the temperature of absorbent in order to get high regeneration efficiency [10]. Propylene carbonate (PC) is another organic solvent, which is readily available and is classified as nonhazardous, and it is often used to remove CO_2 and H_2S from compressed natural gas. The process based on PC is known as Fluor solvent process. It has been reported that PC has a higher CO_2 capacity, less cost compared to water absorption and is easier to regenerate completely compared to DEPG [11]. However, the application of PC in biogas upgrading has been rarely reported [11].

Recently, ionic liquids (ILs) have shown great potential to be used as liquid absorbents for CO_2 removal from different gas streams due to their favorable properties of nonvolatility, thermal stability and high acid gas solubility [12–15]. These unique properties of ILs provide the feasibility to reduce the thermal energy demand for solvent regeneration and the environmental pollution. However, the high production cost, the high viscosity, and the potential toxicity [16] for most of the synthesized ILs limit their industrial applications. As a new type of ILs, deep eutectic solvents (DESs) maintain most of the favorable properties of conventional ILs but avoid the economic and environmental problems. This is because that it is easy to prepare DESs and requires no further purification steps leading to low cost, and most of DESs are biodegradable [15,17–26]. Using DESs as CO_2 absorbents was firstly emerged in 2008 by Li et al. [27]. Following this, plenty of work came out [15,22,26,28–31]. Among the synthesized DESs, choline chloride/urea (ChCl/Urea) is considered as one of the promising solvents to achieve large-scale applications. ChCl/Urea (1:2 on a molar basis) consists of natural compounds, i.e. choline chloride and urea, and thus it is easily biodegradable and with low toxicity. Li et al. [27] and Leron et al. [32] reported the CO_2 solubility in ChCl/Urea (1:2). Their results reveal that ChCl/Urea (1:2) is a promising absorbent for CO_2 absorption, however, its viscosity is much higher than the conventional organic solvents [20]. It has been reported that the addition of H_2O as a co-solvent can decrease the viscosity greatly [26] but still can maintain the high CO_2 capacity, making aqueous ChCl/Urea (1:2) a promising CO_2 absorption solvent.

Therefore, water, DESs-based solvents (e.g. aqueous ChCl/Urea), and organic physical solvents including DEPG and PC can all be used for biogas upgrading or CO_2 separation from other gas streams. These solvents show different behaviors with respect to CO_2 solubility and selectivity as well as other properties such as viscosity, surface tension and density. It has been reported that the higher selectivity can compensate the slightly lower solubility [33]. The viscosity, surface tension and density are also related to the mass transfer rate and then further affect the performance when using these solvents in a traditional mass transfer equipment in the future. This makes it hard to tell which solvent will perform better for biogas upgrading based on the properties only. In addition, it is desirable to know how these properties affect the performance of biogas upgrading, providing knowledge to design new solvents. To address this, process simulation can be used to evaluate and compare the performance of such solvents [13].

Research work has been carried out to compare the performance of solvents for CO_2 separation based on conceptual process. However, the work based on ILs or DESs is very limited probably due to the difficulty in describing the properties and phase equilibria for these complex systems. For example, Shiflett et al. [34] simulated a CO_2 capture process with pure IL and claimed a 16% and 11% reductions in the energy utilization and investment, respectively, compared to the aqueous amine scrubbing. Basha et al. [35,36] developed a conceptual