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Simulation study of deep eutectic solvent-based biogas upgrading process integrated with single mixed refrigerant biomethane liquefaction

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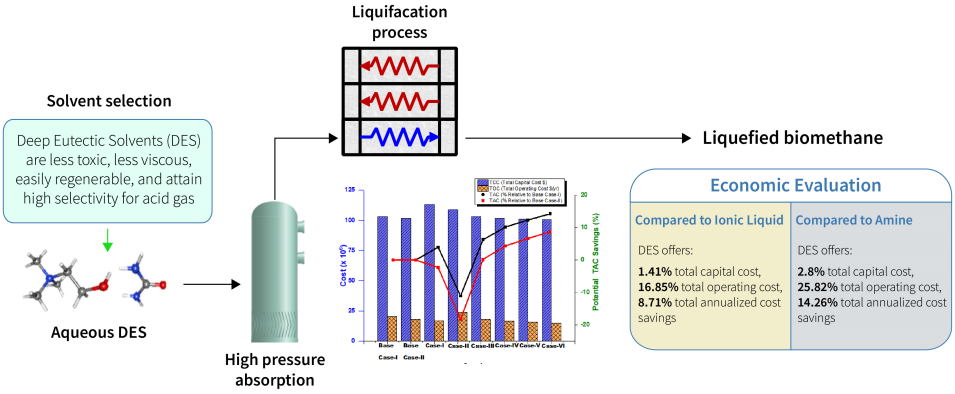
HIGHLIGHTS

- Biogas upgrading integrated with biomethane liquefaction was investigated.
- ChCl/urea was evaluated for biogas upgrading at relatively high pressure.
- The 70% DES led to biomethane with ≥99.0 wt% purity and ≥97.0 wt% recovery.
- The upgraded biomethane was liquefied through a single mixed refrigerant process.
- Biomethane liquefaction process consumed 0.43 kW/kg-biomethane.

GRAPHICAL ABSTRACT



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ABSTRACT

Deep eutectic solvents (DESs) comprise ChCl/urea, in combination with water, have been considered in removing acid gases (CO_2 and H_2S) from biogas. The evaluation of DES for biogas upgrading at relatively high pressure (i.e., >8.0 bar) has not been reported before. The aqueous DES performance has also not been analyzed compared to conventional amines-based solvent (MEA) and ionic liquid (IL). To the best of our knowledge, this is the first study that presents the integration of DES-based biogas upgrading with a mixed refrigerant liquefaction process to facilitate the safe and economical transportation of biomethane over long distances. The biogas considered in this study consisted of 60% CH_4, 39% CO_2, and 1% H_2S. The aqueous ChCl/urea (70 wt%) results in biomethane with ≥99.0 wt% purity and ≥97.0 wt% recovery. Then, this biomethane was liquefied with ≥90% liquefaction rate. Based on the results obtained herein, overall capital, operating, and total annualized cost savings of 2.8%, 25.82%, and 14.26% were achieved using the 70% DES-based integrated process in comparison with the MEA-based integrated process. Whereas 1.41%, 16.85%, and 8.71% capital, operating, and total annualized costs could be saved in comparison with the IL (i.e., [Bmim][PF_6])-based integrated process. It could be deduced that the overall cost of the biomethane value chain can be reduced using the proposed approach.

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Abbreviations and Nomenclatures

$C^{o}_{p,i}$	Purchased cost
AB	Absorber column
[Bmim][PF_6]	1-butyl-3-methylimidaolium hexafluorophosphate
C_{BM}	Bare module cost
ChCl	Choline chloride
CHX	Cryogenic heat exchanger
DEPG	Dimethyl ether polyethylene glycol
DES/F	Deep eutectic solvent to feed ratio
DESs	Deep eutectic solvents
EOS	Equation of state
$F_{BM,i}$	Bare module factor
FD	Flash drum
GHG	Green House Gas
ILs	Ionic liquids
JTV	Joule Thomson valve
LBM	Liquefied biomethane
MEA	Monoethanolamine
MITA	Minimum internal temperature approach
MR	Mixed refrigerant
PC	Propylene carbonate
PR	Peng Robinson
RB	Regenerator column
SMR	Single mixed refrigerant
TAC	Total annualized cost
TCC	Total capital cost
TOC	Total operating cost
TV	Throttling valve
VLE	Vapor-liquid equilibrium

1. Introduction

Fossil fuels account for a significant proportion of global energy, delivering around 80% of the energy worldwide (Qyyum et al., 2020). Fossil fuels are also major contributors to air pollution and greenhouse gas (GHG) emissions. GHGs, especially CO_2, play a primary role in climate change, and it is projected that the concentration of CO_2 will increase by 478–1100 ppm by the year 2100 (IPCC, 2018), which will lead to an increase in the overall environmental temperature of 2–5 °C. Global concerns about addressing climate change were highlighted in the Paris Agreement (UNFCCC, 2015), where it was proposed that the signatories should make efforts to limit the increase in the average global temperature to less than 2 °C, similar to the pre-industrial age, and take measures to restrict the increase in the temperature to 1.5 °C above that of the industrial era (Zain and Mohamed, 2018). Hence, there is an imperative need to find green, cost-effective, alternative energy sources such as biomethane to tackle global warming issues following the Paris Agreement (UNFCCC, 2015).

Biomethane is obtained from biogas upgrading. Biogas comprises CH_4 (50–75%), CO_2 (15–25%), H_2S (<2.0%), and traces of H_2, H_2O, and O_2, depending on the feedstock and operating conditions used (Cucchiella et al., 2018; Scarlat et al., 2018; Tabatabaei et al., 2020). Biogas should be cleaned and upgraded efficiently to obtain biomethane with a purity of ≥97% to overcome the dependency on conventional natural gas (Carnevale and Lombardi, 2015). Therefore, biogas upgrading is one of the major components of the biomethane value chain. The end use of biomethane is critically affected by the efficiency of the upgrading technology. Biogas upgrading through the absorption approach is considered a well-established technology (Hashemi et al., 2019). High biomethane purity with low methane loss is the major feature of the absorption-based biogas upgrading approach and depends on the solvent, and the operating conditions utilized (Qyyum et al., 2020). Considering the environmental, operational, and economic factors, selecting an appropriate solvent is critical for optimizing the absorption-based biogas upgrading process.

Conventionally, amine-based solvents are known to remove acid gases from biogas and natural gas. Monoethanolamine (MEA) is the most well-known first-generation amine-based solvent. The major advantages of MEA are its ideal kinetics, reasonable stability, low capital cost, and high chemical reactivity with acid gases, especially CO_2 (Vega et al., 2018; Augelletti et al., 2020). Although MEA has shown promising performance for acid gas capture in different process plants (including biomethane production plants), many critical issues such as low absorption capacity, high enthalpy of reaction, thermal degradation, highly corrosive degradation products, and high operating costs for solvent regeneration are associated with the use of MEA (Vega et al., 2018; Haider et al., 2020).

To solve the issues related to typical amine-based solvents, ionic liquids (ILs) are emerging as a new class of solvents for acid gas removal applications. As their dominant advantages, ILs are non-toxic, non-flammable, biodegradable, recyclable, environmentally-friendly, have high chemical and thermal stability, are accessible, and favorably absorb various species for a wide range of applications (Lei et al., 2014; Xu et al., 2015; Nematollahi and Carvalho, 2019). The reaction enthalpy of ILs can be tuned by varying the anion group, and such flexibility in the structure of ILs can diversify their applications. However, ILs are costly, and this is a significant hurdle for their large-scale commercial applications. Deep eutectic solvents (DES) have been introduced and considered as another class of ILs (Alkhatib et al., 2020; Słupek et al., 2020). Although DESs have the same characteristics as ILs, the price is significantly lower than ILs (Pena-Pereira and Namieśnik, 2014; Kamgar et al., 2017), making them more attractive for large-scale applications. Among the various available DESs, choline chloride/urea (ChCl/urea; 1:2 by mole) is one of the most suitable candidates for commercial-scale biogas upgrading (Ma et al., 2018a) for its relatively low toxicity and highly biodegradable nature. The fundamental parameters for the selection of the aqueous DES (ChCl/Urea + water) are compared with those of pure DES, aqueous MEA, and a pure IL, i.e., [Bmim]PF_6 (recently studied (Haider et al., 2019) for biogas upgrading), in **Table 1**.

Ma et al. (2018a) surveyed and evaluated the thermophysical properties of pure and aqueous ChCl/Urea for large-scale biogas upgrading. By implementing a database for pure and aqueous ChCl/Urea in Aspen Plus, they compared the performance of aqueous ChCl/Urea *vs.* pure water for biogas upgrading. In another study, Ma et al. (2018b) compared the performance of aqueous (ChCl/Urea) with those of three physical solvents (water, dimethyl ether of polyethylene glycol (DEPG), and propylene carbonate (PC)) for biogas upgrading, and highlighted the possible techno-economic benefits of using the DES. They used Aspen Plus to evaluate the biogas upgrading processes and concluded that when the biogas production capacity was higher, pure water and aqueous DES performed better than

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Table 1.
Comparison of DES with MEA and IL.

Parameters	$(Pure)_{DES}$	$(Aq.)_{DES}$	$(Aq.)_{MEA}$	IL	Reference
Selectivity (H_2S/CO_2)	5.5-4.76	2.9-3.1	2.6-4.0	1.2-3.7	Dubois and Thomas (2010); Hadj-Kali et al. (2020); Handy et al. (2014); Kazmi et al. (2019); Liu et al. (2019)
Selectivity (H_2S/CH_4)	60.8-42.8	43.7-35.4	-	-	
Selectivity (CO_2/CH_4)	11.1-9.1	15.2-12.4	156.72	38.24	
Toxicity (LC_{50} / EC_{50})	>100 mg/L	>100 mg/L	7100 mg/L	634 mg/L	Azimova et al. (2009); Juneidi and Hayyan (2016)
Viscosity (cP)	750	6.0	2.48	376	Amundsen et al. (2009); Drahansky et al. (2016); Wang et al. (2012)
Corrosivity (m/y)	negligible	1×10^{-5}	2.43	negligible	Barbosa et al. (2020)

DPEG. Total annualized cost (TAC) savings of 30% and 45% were achieved by using PC and aqueous DES compared to water. However, in both studies by Ma et al. (2018 a and b), the analyses were performed at low pressure (i.e., 8.0 bar); which was significantly lower than the critical pressure of biomethane. Pressure has a significant impact on physical absorption and biomethane liquefaction, which is considered the most economical and safe approach for biomethane transportation *via* cargo trucks or ships. Although biomethane liquefaction itself is regarded as an energy-intensive process, if the pressure is near or greater than the critical pressure of biomethane, the liquefaction process is favorable as it can achieve an overall low power consumption (Rehman et al., 2020).

Liu et al. (2019), Xie et al. (2016), Leron et al. (2013), Li et al. (2008), Mirza et al. (2015), and Hsu et al. (2014) published the solubility data for the $CH_4/CO_2/H_2S$ system in pure and aqueous ChCl/Urea, where the data were acquired at 30–80 °C and 0.105–45.04 bar. By using these solubility data, biogas upgrading systems can be designed and analyzed at high pressure (i.e., >8.0 bar) by considering biomethane liquefaction for economical and safe transportation. Therefore, this study is the first to investigate biomethane liquefaction from the process engineering perspective and biogas (consisting of CH_4, CO_2, and H_2S) upgrading using DES at high pressure (i.e., 36 bar). This article is structured as follows: the process description and simulation basis are described in *Section 2*. *Section 3* presents process analysis in terms of the optimal design variables, parameters, constraints, and energy requirements. *Section 4* offers an economic evaluation of the proposed integrated process for the preliminary determination of commercial feasibility. Finally, the conclusions and future work are covered in *Section 5*.

2. Research Methodology: proposed process

2.1. Process description

The proposed process comprises two major sections: biogas upgrading using a DES and biomethane liquefaction using a single mixed-refrigerant (SMR) process. The schematic presentation of the process and the main parameters (temperature, pressure, and flowrate) are shown in **Figure 1.** A biogas stream (S2) (at 30 °C and 1 bar pressure) from an anaerobic digester (66759 $m^3 h^{-1}$) was compressed through multistage compression at 36 bar pressure and introduced into an eight-stage vertical absorption column (AB-1). An aqueous solvent stream (S1) was introduced from the top of the column (AB-1) at 30 °C and 36 bar pressure; the stream flows downwards and interacts with the upward flowing biogas. During this process, the biogas is upgraded by selective adsorption of CO_2 and H_2S, with a biomethane recovery of ≥97 wt% and 99 wt% purity. The bottom stream (S4), rich in the solvent, H_2S, and CO_2, is discharged to the flash column (FD-1), which operates at a 13.2 bar pressure to recover and further purify CH_4, which is recycled into the absorption column. The pressure of the bottom stream from FD-1 is further reduced to 1 bar through the valve TV-1, and this stream is introduced into the regenerator column (RB-1) from the top, while the compressed air stream is introduced from the bottom of the column using the blower (B-1). Both streams interact counter-currently to regenerate the solvent mixture, which is recycled (S8) to the absorber using the pump (P-1).

The purified biomethane gas (99 wt%) at a temperature of 30 °C and 36 bar pressure is fed to the liquefaction section parallel to the refrigerant stream (S26), which comprises a mixture of nitrogen, methane, ethane, propane, and *i*-propane at 40 °C in a cryogenic heat exchanger (CHX-1). The refrigerants are compressed in multistage compressors with an inter-stage cooling system to raise the pressure to 52.5 bar. Subsequently, the high-pressure mixed refrigerant (MR) is expanded using a Joule-Thomson valve (JTV-1), which expands the mixture to the optimal evaporating pressure of 3.74 bar, resulting in a temperature drop to -149.71 °C. This expanded MR is recycled (S12) into the cryogenic heat exchanger (CHX-1), which exchanges the latent heat of vaporization from the incoming biomethane (S3) and warm MR (S26) stream. Finally, the sub-cooled liquefied biomethane (LBM) stream (S13) at -159.47 °C and 1.209 bar are sent to the storage unit. The super-heated saturated vapor stream of MR (S27) at a temperature of 38 °C is recycled to the compression unit, where the MR is introduced into a series of multistage compression units. The system is equipped with inter-stage cooling at each stage, where the intercooler outlet temperature is set to 40 °C.

2.2. Process simulation

To analyze any processes through modeling and simulation, the thermodynamic properties, including the binary interaction parameters, enthalpy, and entropy, are critical factors that determine the process reliability. The Aspen Plus simulator has an extensive and wide-range database with versatile thermodynamic fluid packages. Therefore, the process proposed in the present study was simulated by using Aspen Plus® v10. However, the solubility data for the studied system is not available in Aspen's database so far. Thus, the solubility data for CO_2, H_2S, H_2O, and CH_4 in ChCl/Urea (given in **Table 2**) were validated by performing rigorous regression using the Peng Robinson (PR) equation of state (EOS). The selected EOS model is distinct from most other enhanced models in the Aspen database as it can handle a broader range of temperatures and pressures and has the largest database of binary interaction parameters for a wide range of components (Nawaz et al., 2019).

Hence, by correlating the vapor-liquid equilibrium data using PR, the obtained binary interaction parameters after regression have minimum deviation, which shows a good agreement with experimental results, as shown in **Table 3**. The estimated solubilities of the respective components achieved by regressing the vapor-liquid equilibrium (VLE) data are shown in **Figures 2a**, **b**, and **c** for CO_2, H_2S, and CH_4 in ChCl/Urea, respectively.

The proposed process and base cases were simulated based on the following fundamental assumptions:

- Heat loss to surroundings was assumed to be zero
- Minimum pressure drop of 0.1 bar across inter-stage coolers
- 75% isentropic efficiency was maintained for each compressor and pump
- Minimum internal temperature approach (MITA) is fixed at 2.0 °C for efficient heat exchange through a multistage biomethane liquefaction cryogenic heat exchanger.

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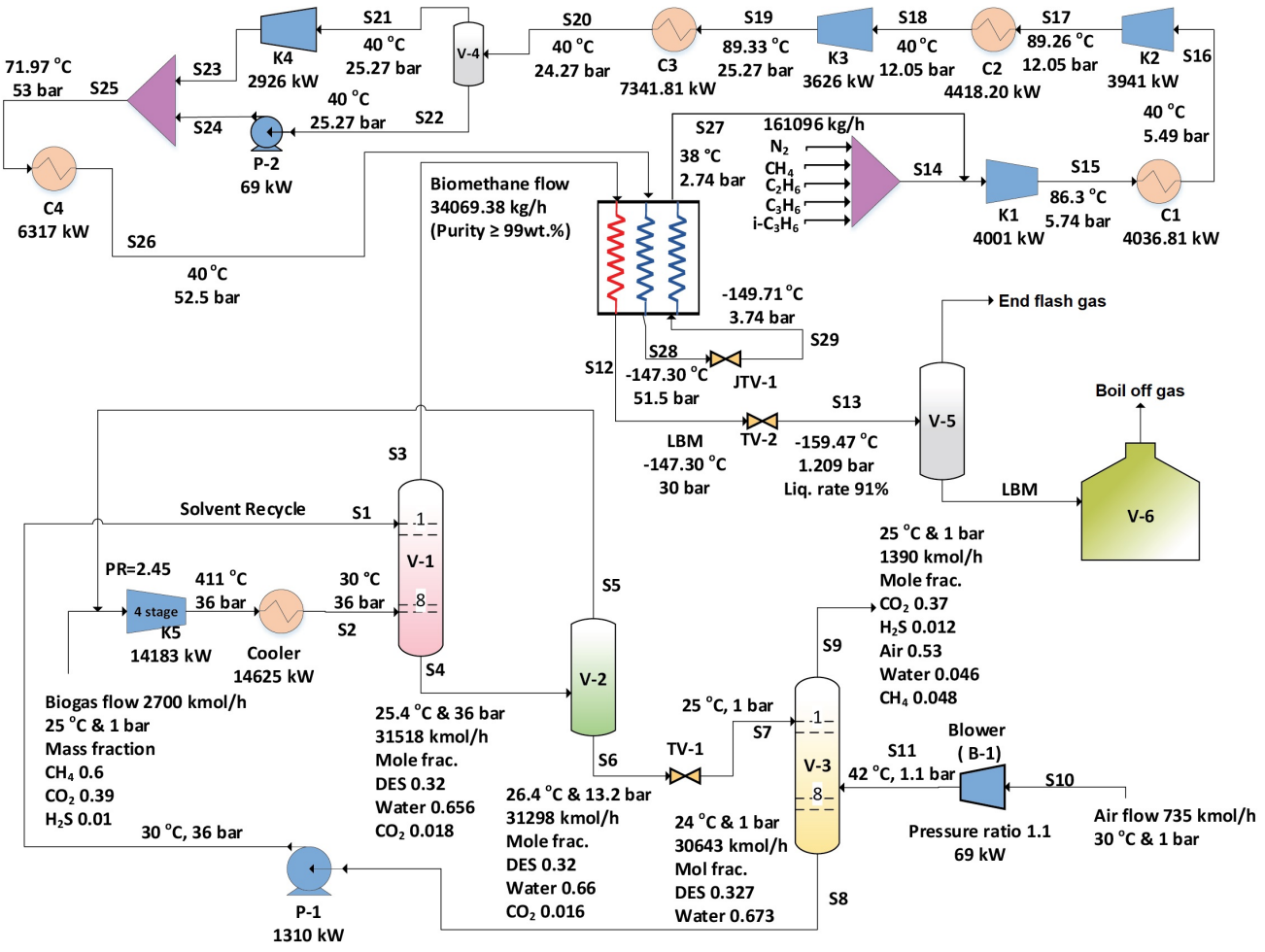

Fig. 1. Biomethane liquefaction followed by DES-based biogas upgrading. LBM: Liquefied biomethane.

Table 2.
Vapor-liquid equilibrium (VLE) data for CO_2, CH_4, and H_2S and H_2O in ChCl/Urea.

DES	System	T (°C)	P (bar)	Solubility range	No. of points	Reference
ChCl/Urea (1:2)	CO_2 +H_2O	40.05	0.301-8.096	0.038-0.111	30	Hsu et al. (2014)
		60.05	0.54-9.246	0.032-0.107	33	
		80.05	0.344-8.757	0.029-0.097	28	
	CO_2	40	11.3-125	0.077-0.309	8	Li et al. (2008)
		50	10.1-111	0.060-0.271	8	
		60	10-127.3	0.057-0.270	8	
	CO_2	35.05-55.05	6.51-45.04	0.042-0.195	15	Xie et al. (2016)
	CH_4	35.05-55.05	5.48-36.18	0.012-0.089	15	
	CO_2	40.05-80.05	0.105-2.022	0.00038-0.01624	20	Liu et al. (2019)
	CH_4	40.05-80.06	0.106-2.028	0.00004-0.00110	20	
	H_2S	40.05-80.07	0.101-2.021	0.00151-0.04640	20	
	CO_2	35.85-55.85	0.405-1.535	0.000906-0.0035	21	Mirza et al. (2015)
	CO_2	30-70	2.99-59.11	0.1562-3.5592	34	Leron et al. (2013)

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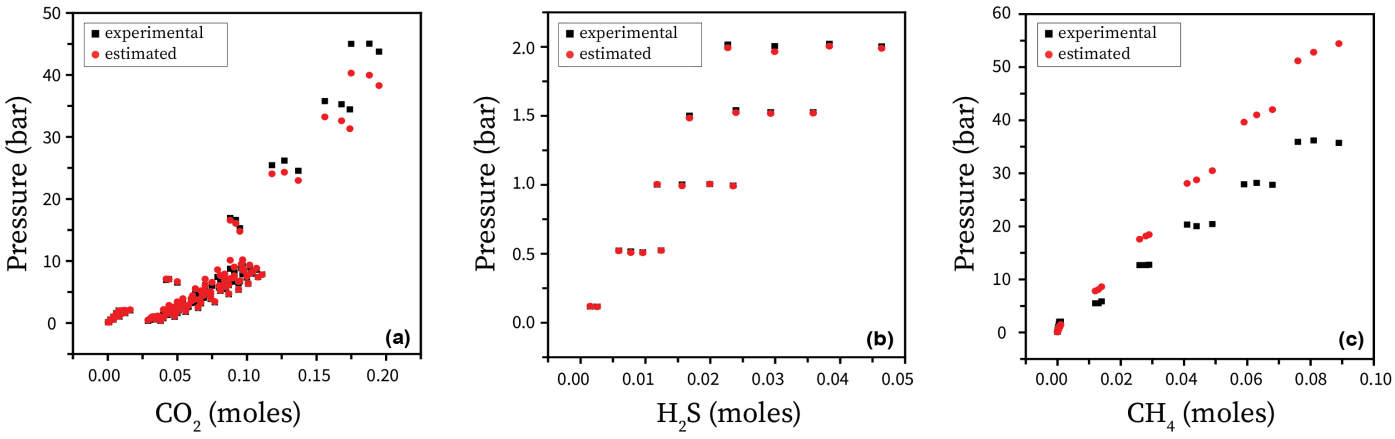

Fig. 2. Solubility of **(a)** CO_2, **(b)** H_2S, and **(c)** CH_4 in ChCl/Urea.

Table 3.
Binary interaction parameters.

Peng Robinson-based binary parameters				
i	***j***	**KA_{ij}**	**KB_{ij}**	**Deviation**
ChCl/urea	CH_4	0.030768859	2.28E-05	0.030892042
ChCl/urea	CO_2	0.058406364	7.27E-06	0.028740977
ChCl/urea	H_2S	-0.13458251	0.000568031	0.132269889
ChCl/urea	H_2O	-0.024105302	0.002267954	0.027514354
Average deviation				**0.055**

3. Results and Discussion

3.1. Process analysis: optimal design variables and parameters

The proposed integrated process was investigated and analyzed by varying the concentration of the DES solvent, as shown in **Figure 3**. **Table 4** tabulates the design variables, parameters, constraints, and overall energy consumption for all the investigated cases compared to MEA- and IL-based biogas upgrading.

The effect of varying the concentration of the aqueous solutions of DES on biogas upgrading was analyzed using the proposed configurations. It was reported that the addition of 30% water to DES lowered its viscosity by 90% (Zhang et al., 2012; Kalhor and Ghandi, 2019). Therefore, the analysis included six cases in which the solvent's composition was varied; the data were compared with those of alkanol amine- and IL-based solvents.

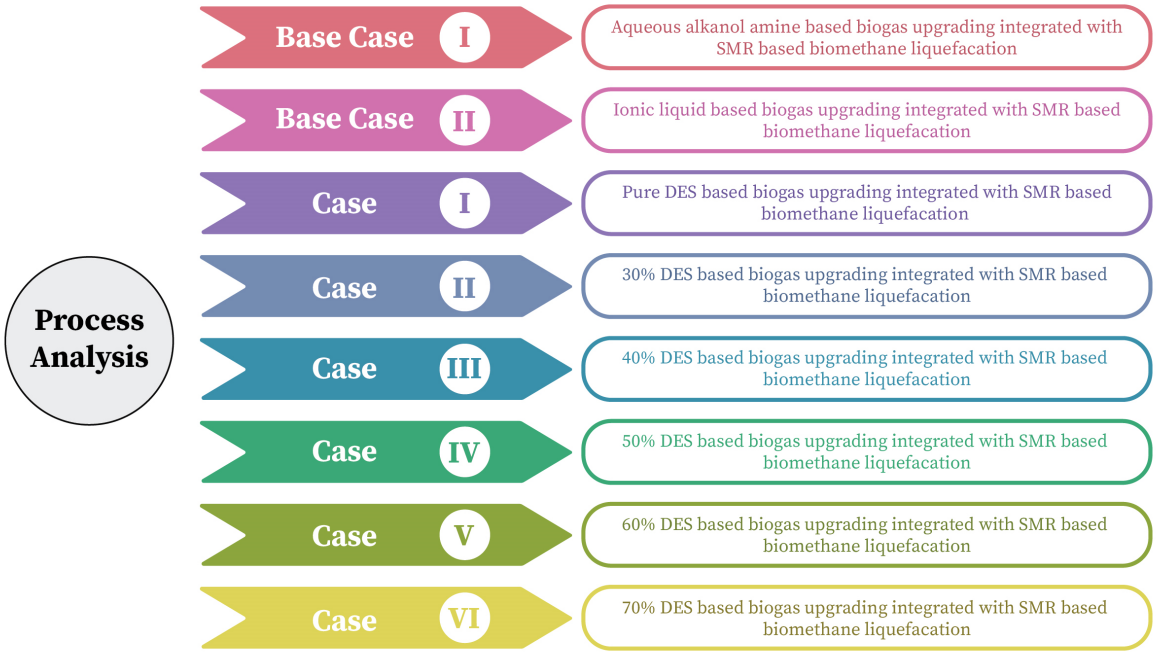

Fig. 3. Process analysis cases for DES-based biogas upgrading integrated with biomethane liquefaction.

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Table 4.
Design variables and parameters for biomethane liquefaction followed by DES-, MEA-, and IL-based biogas upgrading.

Variables/Parameters	Base case-I	Base case-II	Case-I	Case-II	Case-III	Case-IV	Case-V	Case-VI
Biogas upgrading								
Solvent composition (wt%)	MEA (30), H_2O (70)	[Bmim][PF_6] (100)	ChCl/urea (100)	ChCl/urea (30), H_2O (70)	ChCl/urea (40), H_2O (60)	ChCl/urea (50), H_2O (50)	ChCl/urea (60), H_2O (40)	ChCl/urea (70), H_2O (30)
Constraints								
Purity of biomethane (wt%)	0.99	0.99	0.99	0.99	0.99	0.99	0.99	0.99
Recovery of biomethane (wt%)	0.99	0.96	0.97	0.97	0.97	0.97	0.97	0.97
H_2S reduction (ppm)	≤ 10	≤ 10	≤ 10	≤ 10	≤ 10	≤ 10	≤ 10	≤ 10
CO_2 removal (%)	0.99	0.985	0.99	0.99	0.99	0.99	0.99	0.99
Absorber								
Flowrate of biogas (kg/h)	58000.0	58000.0	58000.0	58000.0	58000.0	58000.0	58000.0	58000.0
Flowrate of biogas (m^3/h)	66759	66759	66759	66759	66759	66759	66759	66759
Flowrate of solvent (m^3/h)	1181.63	909.463	1168.2825	12083.379	5888.14	3137.673	1902.6315	1134.903
Solvent/Feed ratio (m^3/m^3)	0.0177	0.01362	0.0175	0.181	0.0882	0.047	0.0285	0.0166
Absorber stages	20	14	10	14	12	10	8	8
Absorber pressure (bar)	36	36	36	36	36	36	36	36
Absorber temperature (°C)	40	30	30	30	30	30	30	30
Flash drum pressure (bar)	-	-	6.8	14.7	14.7	14	12.8	13.2
Regenerator								
Regenerator stages	19	1	8	8	8	8	8	8
Regenerator temperature (°C)	40	80	40	40	40	40	40	40
Regenerator pressure (bar)	1.1	0.1	1.1	1.1	1.1	1.1	1.1	1.1
Reflux ratio	2	-	-	-	-	-	-	-
Duty of regenerator (kW)	26718	4594.41	-	-	-	-	-	-
Airflow rate (m^3/h)	-	-	18777.8	55429	40893.6	31055.8	24952.9	17482.4
Air/DES rich ratio (m^3/m^3)	-	-	0.282	0.825	0.615	0.469	0.377	0.257
Power of pump for recycling (kW)	469.7	1359.11	1686.74	16245.3	6877.37	4244.72	2590.02	1306.94
Power of air compressor (kW)	-	-	67	219	161	111	89	69
Specific thermal load (kWh/kmol biogas)	5.067	5.067	6.369	6.146	5.244	5.24	5.29	5.76
Specific thermal load (kWh/kg biogas)	0.235	0.235	0.296	0.286	0.244	0.244	0.246	0.245
Biomethane liquefaction								
Constraints								
LNG product (liquid fraction)	0.91	0.91	0.91	0.91	0.91	0.91	0.91	0.91
MITA (°C)	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0
Design variables								
Mass flow rate of nitrogen, m_{N2} (kg/h)	9357.5	9357.5	9357.5	9357.5	9357.5	9357.5	9357.5	9357.5
Mass flow rate of methane, m_{C1} (kg/h)	39561.5	39561.5	39561.5	39561.5	39561.5	39561.5	39561.5	39561.5
Mass flow rate of ethane, m_{C2} (kg/h)	17464.4	17464.4	17464.4	17464.4	17464.4	17464.4	17464.4	17464.4
Mass flow rate of propane, m_{C3} (kg/h)	50397.2	50397.2	50397.2	50397.2	50397.2	50397.2	50397.2	50397.2
Mass flow rate of i-pentane, m_{iC5} (kg/h)	44315.6	44315.6	44315.6	44315.6	44315.6	44315.6	44315.6	44315.6
Refrigerant low pressure (bar)	9.3	9.3	9.3	9.3	9.3	9.3	9.3	9.3
Refrigerant discharge pressure (bar)	2.74	2.74	2.74	2.74	2.74	2.74	2.74	2.74
Pressure ratio	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1
Objectives								
Total compression power (kW)	14563	14563	14563	14563	14563	14563	14563	14563
Specific (molar) compression power requirement (kW/kmole biomethane)	6.89	6.89	6.89	6.89	6.89	6.89	6.89	6.89
Specific (mass) compression power requirement (kW/kg biomethane)	0.43	0.43	0.43	0.43	0.43	0.43	0.43	0.43

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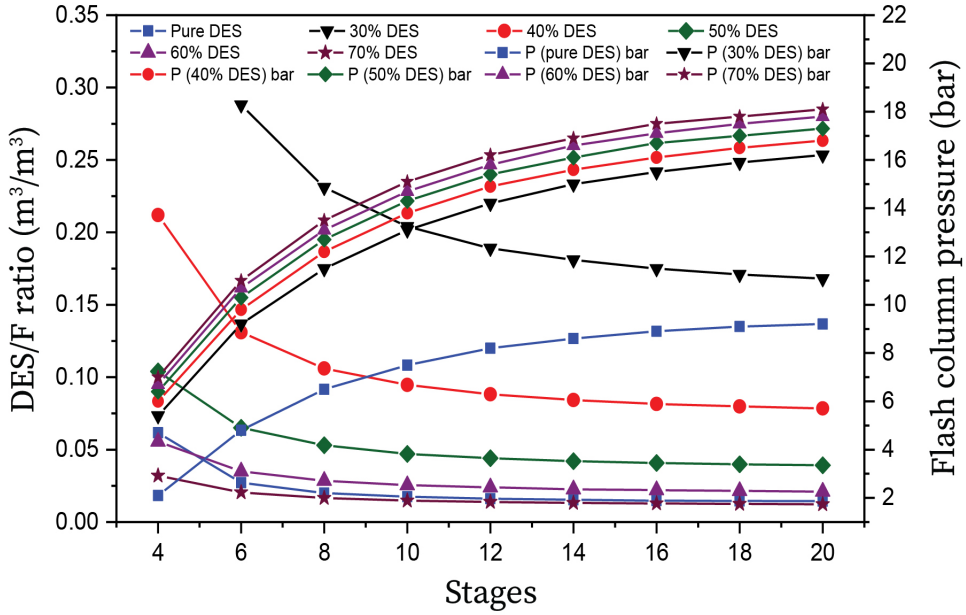

Fig. 4. Absorber analysis based on stages for selection of DES flow and flash drum pressure.

Consequently, a sensitivity analysis was carried out for the upgrading section to evaluate the required solvent/feed ratio when the number of stages was varied to achieve constraint satisfaction. The optimum solvent flow rate associated with the absorber stages is pivotal as it is directly associated with the solvent recovery unit. Notably, a high solvent flow rate during the absorption process leads to cost-intensive recovery in the solvent regeneration step for the high power required to pump back the solvent and the high flow of air required to regenerate the solvent. **Figure 4** shows the DES's estimated flow regarding the absorber stages for the required purity and recovery of biomethane. The analysis was performed for aqueous DES with various compositions, and the deep eutectic solvent to feed (DES/F) ratio was calculated to estimate the number of stages in the absorber. The addition of water reduces the solvent's viscosity but increases the overall flow of the DES in the absorber to meet the process constraints. It was predicted that after a certain stage number, the DES/F ratio would become constant in each case. Hence, the process configurations proposed at the stages required for cases I–VI were 10, 14, 12, 10, 8, and 8, respectively; the corresponding DES/F was 0.0175, 0.181, 0.0882, 0.047, 0.0285, and 0.0166, respectively.

The flash column connected to the absorber enhances the purity and recovery of biomethane in the top stream. Nevertheless, optimizing the pressure in the flash column is a crucial factor. The flow of the recycle stream changes as the pressure of the flash column varies. Thus, reducing the flash column pressure directly affects the power of the biogas compressor required to accelerate the ejection of the gas from the flash drum. Notably, the compressor power needed to recycle the vapor stream of the flash column was the lowest with the use of 70% DES as the pressure reduction in the flash column was about 13.5 bar. Whereas the pressure in the flash column was lower in the other cases, as shown in **Figure 4**. The top stream from the absorber containing biomethane is sent to the separate section where liquefaction takes place. In contrast, the bottom stream from the absorber is rich in DES that must be recovered in the solvent recovery column.

Air stripping was used to recover and purify the solvent by separation from the acid gases in the solvent regeneration column. The air was compressed at 1.1 bar through the mechanical blower and introduced into the regenerator, where the acid gas components were separated from the DES through air stripping. The flow of air for each selected stage was set such that DES was recovered with 99.99% purity and recycled to the absorption column by pumping at 36 bar. The power required for the air blower depends on the airflow rate and regenerator stages, as shown in **Figure 5**. Accordingly, the power required to compress the air is reduced as the number of stages in the regenerator increases and becomes constant at the 8th stage, given that low airflow is required to recover the DES from acid gas. Therefore, for 70% DES, eight stages were selected for the regenerator for solvent recovery. Similarly, for the other cases, the Air/DES ratio became constant at the 8th stage. Nevertheless, the airflow was comparatively high compared to that required for 70% DES, where the former ultimately requires greater compression power and causes the overall cost of the process to increase.

The biomethane stream obtained from the top of the absorber was liquefied *via* the SMR process. Therefore, the parametric analysis of the liquefaction section was carried out to determine the optimal design objectives. The optimal design parameters and variables are listed in **Table 5**. There are two process streams in the liquefaction section, one is the refrigeration loop, and the second is the NG stream, which is passed through the cryogenic heat exchanger and liquefied. The refrigerant mixture enters the refrigeration loop at 2.74 bar, and 40 °C at a flow rate of 161096.3 kg h^{-1} and is circulated in the refrigeration cycle. Optimizing the refrigerant flow rate is crucial for the liquefaction process as this parameter is directly related to the compression power. Therefore, the refrigerant flow rate and liquefaction rate were assumed as process variables. The refrigerant mixture comprises nitrogen, methane, ethane, propane, and *i*-pentane, depending on liquefaction requirements. The mixture of low and high boiling solvents is typically used for precooling and liquefaction, where at the optimal flow rates of 9357.5, 39561.5, and 17464.4 kg h^{-1} for nitrogen, methane, and ethane, respectively, the mixture has the refrigeration capacity to produce LNG. On the other hand, a high flow rate of propane and *i*-pentane is useful for precooling the refrigeration mixture. Furthermore, a high flow of *i*-pentane (44315.6 kg h^{-1}) is used because this compound has a high refrigeration capacity, thereby reducing the overall compression power. The inter-stage cooling system was implemented during compression to maintain a stable refrigerant temperature. In this context, the design parameters would have maintained the pressure drop at the coolers at 0.25 bar; the temperature at the outlet after every compression and the corresponding cooling stage was 40 °C. Notably, after the third cooling stage, some refrigerant mixtures were converted into liquid fractions (0.12) because of the high-boiling *i*-pentane. Thus, the mixture was separated in the flash tank and compressed in the respective process units and mixed again to obtain a unified cooling stream. The minimum internal temperature approach (MITA) inside the cryogenic heat exchanger was defined as a process constraint and fixed at 2.004 °C. Conclusively, the liquefaction ratio based on the MITA value was 0.91, and the pressure ratio was 2.1 in the multistage compressor. Furthermore, the overall compression

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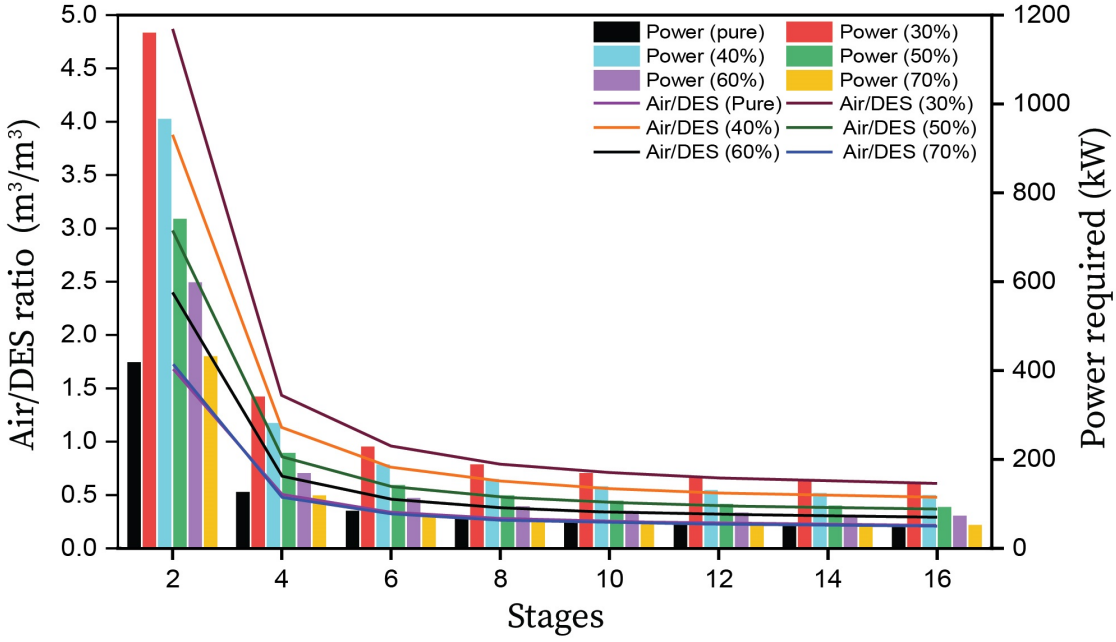

Fig. 5. Regenerator analysis to choose airflow rate based on absorber stages and power required in an air blower.

Table 5.
Capacity parameters and values of constants for equipment used in this study (Turton et al., 2008).

Equipment category	K_1	K_2	K_3	Capacity, Unit
Absorber	3.4974	0.4485	0.1074	Volume, m^3
Packing	2.4493	0.9744	0.0055	Volume, m^3
Regenerator	3.4974	0.4485	0.1074	Volume, m^3
Air cooler	4.0336	0.2341	0.0497	Area, m^2

power required to compress the mixture was 14563 kW, and the specific compression power was 6.89 kW/kmole biomethane.

4. Economic evaluation

Energy efficiency-based optimization of the process is one aspect to be considered in process design. Nevertheless, evaluating the economic feasibility is the most essential factor and mainly depends on the TAC, a function of the capital cost (TCC) and operating cost (TOC). For the TAC calculation, a payback period of 5 years was set for all process configurations to keep the fair comparisons among all cases. The cost relation is presented in **Equation 1**.

$$TAC = \left(\frac{Capitalcost}{Paybackperiod}\right) + Operatingcos \quad \text{Eq. 1}$$

The TCC estimation is based on the bare module cost (C_{BM}), which is a function of the bare module factor ($F_{BM,i}$) corresponding to **Equations 2** and **3**, while $C^{\circ}_{p,i}$ is the purchased cost, expressed in **Equation 4** (Turton et al., 2008).

$$FCI = \sum_{i=1}^{N} C_{BM,i} \quad \text{Eq. 2}$$

$$C_{BM,i} = C^{o}_{p,i} F_{BM,i} \quad \text{Eq. 3}$$

$$\log_{10}(C^{o}_{p,i}) = K_1 + K_2 \log_{10}(A_i) + K_3[\log_{10}(A_i)]^2 \quad \text{Eq. 4}$$

where, K_1, K_2, and K_3 denote the selective equipment-based constant parameters, and A is the capacity measure. The parametric relations and constant values were taken from the literature (Turton et al., 2008) and are listed in **Table 5**. The volume of the column was calculated as $\pi D^2 L/4$, and that of the packing as $\pi D^2/4$. In the case of the cryogenic heat exchanger, the plate-fin heat exchanger type was used for the capacity-based small-scale liquefaction plant. However, the value of F_{BM} for this specific heat exchanger is not documented in the open literature. Therefore, the Aspen economic analyzer was used to estimate the capital cost of the cryogenic heat exchanger. Similarly, "UA" was determined from the simulation results, where "U" is the heat transfer coefficient, and "A" is the area with the air cooler. Nevertheless, "A" is the capacity factor for calculation of the capital cost, which was estimated by considering "U" as 568 W $m^{-1}K^{-1}$ for the air-cooler based on the literature (*The Basics of AIR-COOLED HEAT EXCHANGERS. HUDSON Products Corporation, 2007*).

The capacity range for pumps documented by Turton (Turton et al., 2008) was up to 300 kW, while in the current study, even a higher power was required for the pumps to operate on the viscous DES. Therefore, the capital cost relations for the high pressure and high capacity data were taken from the *Engineering* design book by Douglas (Erwin, 2015). The cost relation is presented as **Equation 5**, in which F_p denotes the pressure factor, and F_M represents the material factor. Similarly, the compressors' capital cost was estimated by applying **Equation 6**, in which M & S represents the Marshall and Swift Index; W is the power of the compressor in *hp*; and F_c is the correction factor, where the value is unity.

$$Base\ module\ unit\ cost = \text{Base cost} \times (F_p + F_M) \times Cost\ Index \quad \text{Eq. 5}$$

$$Compressor\ (CC) = 517.5 \times \left(\frac{M\ \&\ S}{280}\right) \times W^{0.82} \times F_c \quad \text{Eq. 6}$$

The TOC of the process was calculated by considering the electricity cost for power generation required to compress the biogas, air compression

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for regeneration of the solvent, pumping cost for solvent recycling in the upgrading section, and compression of the refrigerants for liquefaction. The major portion of the power required in each section was due to the compression of the biogas and refrigerants. The cost of electricity was assumed to be \$16 GJ^{-1}, leading to a cost of \$504.5 kWh^{-1} (Turton et al., 2008). **Table 6** lists the equipment for the upgrading and liquefaction section and the corresponding power consumption.

Table 6.
Power required in unit operations.

Equipment	Power (kW)
Biogas upgrading	
Biogas compressor	14183
Air blower	69
DES pump	1307
Biomethane liquefaction	
K-1	4001
K-2	3941
K-3	3626
K-4	2926
P-1	69

Table 7 presents the cost estimate for all the proposed DES-based integrated processes, compared with the MEA- and IL-based integrated processes. The TAC for each case mainly depends on the compressors. However, economic comparison of the MEA-based process with the proposed configurations reveals that for the former, a high cost is incurred for the energy-intensive solvent regeneration, i.e., around USD 6.69 $\times 10^6$. Similarly, the overall operating cost for the amine-based biogas upgrading and subsequent liquefaction was estimated at USD 20.487 $\times 10^6$, which is far greater than that of the DES-based upgrading process. Unlike the amine-based process, the capital cost for the absorber in IL-based biogas upgrading is very high (USD 6.06 $\times 10^6$) because of the viscous and heavy solvent. The prolonged mass transfer rate means that a high flow rate of the IL is required to absorb CO_2. Nevertheless, regeneration is much easier and more energy-efficient for the high thermal stability of ILs, which makes them stable at high temperatures; the total operating cost was calculated at USD 4.03 $\times 10^6$. Hence, the overall cost of IL-based upgrading and liquefaction stands at USD 18.277 $\times 10^6$.

For the DES configurations, the highest TAC of USD 45.709 $\times 10^6$ was calculated for the 30% DES system, where most of the cost was attributed to the high DES rate required in the absorber for compression, and in the intensive regeneration section (similar to the other case). Notably, the data tabulated in **Table 7** show that gradually increasing the amount of water in the DES reduced the overall process cost but was only effective up to 40–70% DES to water. Further addition of water increases the energy consumption for solvent recycling and is thus cost-intensive. Besides, the operating cost savings were mainly reduced for the aqueous solution, which reduces the DES's viscosity.

Table 7 also presents the potential TCC, TOC, and TAC savings using the DES-based integrated process compared to base case-I and base case-II. Accordingly, compared to base case-I, TAC savings of case-I, III, IV, V, and VI were 3.95, 6.24, 10.16, 12.32, and 14.26%, respectively. Case II had a negative potential savings of -11.0438%, suggesting that this case would not be economically beneficial, mainly because of the high DES rate. Similarly, the TAC savings obtained in cases III–VI relative to the IL-based biogas upgrading process were 0.18, 4.35, 6.65, and 8.71%, respectively, because of the energy efficiency in the regeneration section for the DES-based processes compared with the IL-based biogas upgrading process. Cases I and II had negative potential savings of -2.26% and -18.23%, respectively, showing that no cost-savings were incurred in the corresponding cases *vs.* for the IL process.

Hence, based on the process analysis and cost estimation, the 70% DES process was chosen for integration with the liquefaction process. The proposed SMR liquefaction process is cost-intensive in terms of its capital investment as it requires multistage compression of the refrigerants, while a cryogenic heat exchanger is used to liquefy the biomethane. As an added advantage, the proposed scheme provides an overall potential TAC savings of 14.26 and 8.71% compared to the amine- and IL-based configurations. The calculated TCC was USD 68.42 $\times 10^6$, while the operating cost was even lower than that of the upgrading process as biogas is compressed at high pressure (36 bar) before entering the absorber. However, based on the TAC, the unit cost for the upgrading and liquefaction processes was USD 0.6 $kmol^{-1}$ of biogas and \$1.14 $kmol^{-1}$ of biomethane, respectively.

5. Conclusions and future directions

This study investigated the potential of DES for biogas upgrading at 36.0 bar. Different concentrations of DES were investigated for the simultaneous removal of CO_2 and H_2S from biogas. The upgraded biogas (biomethane) was further liquefied by using a single mixed refrigerant comprising nitrogen, methane, ethane, propane, and *iso*-pentane. The following major conclusions could be drawn from the proposed analysis:

- The process of employing DES with 70 wt% concentration could efficiently and economically remove acid gases from biogas.
- DES-based biogas upgrading integrated with SMR-LBM affords 14.26 and 8.71% TAC savings compared to the MEA and IL-based integrated processes, respectively.

Table 7.
Economic evaluation for different cases of DES-based biogas upgrading integrated with liquefied biomethane (LBM) process.

Cost		Base cases		Studied cases					
		Base case-I	Base case-II	Case-I	Case-II	Case-III	Case-IV	Case-V	Case-VI
TCC (USD $\times 10^6$)		103.38	101.92	113.29	108.96	103.22	101.71	101.11	100.48
TOC (USD $\times 10^6$)/yr		20.487	18.277	16.877	23.917	17.947	16.637	15.867	15.197
TAC (USD $\times 10^6$)/yr		41.163	38.661	39.535	45.709	38.591	36.979	36.089	35.293
Potential savings relative to base case-I	**TCC (%)**			-9.58	-5.39	0.15	1.61	2.19	2.80
	TOC (%)			17.62	-16.74	12.39	18.79	22.55	25.82
	TAC (%)			3.95	-11.04	6.24	10.16	12.32	14.26
Potential savings relative to base case-II	**TCC (%)**			-11.15	-6.90	-1.27	0.20	0.79	1.41
	TOC (%)			7.65	-30.85	1.80	8.97	13.18	16.85
	TAC (%)			-2.26	-18.23	0.18	4.35	6.65	8.71

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- It cannot decisively be concluded that DES is an economical option for biogas upgrading. The economic feasibility depends on the concentration of DES, feed biogas composition, and conditions. For example, the TCC for case-I (pure DES) was 9.58% and 11.15% higher (negative savings) than base case-I and II, respectively. Similarly, case-II (30% DES) was totally infeasible based on the TCC, TOC, and TAC, compared to base case-I and II.
- Because biomethane liquefaction is a cryogenic operation, the methane content should be ≥99.99%, with 99.99% recovery. Nevertheless, the proposed schemes failed to yield such high purity and recovery. Therefore, 0.2% CO_2 (by mole) remains in the upgraded biogas, which can lead to undesirable CO_2 freezing in the liquefaction section.

Future works should be focused on determining the TAC savings opportunities in the studied integrated cases, including the base cases, through heat integration of the biogas upgrading and liquefaction section.

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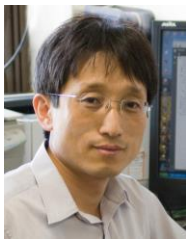

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