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ARTICLE

Removal of CO₂ and H₂S from biogas and enhanced compressed bio-methane gas production from swine manure and elephant grass

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ABSTRACT

Anaerobic co-digestion of concentrated swine manure with elephant grass silage was used, and this study carried out in full-scale three continuously stirred tank reactors was operated at mesophilic temperature. The Biogas impurity removal procedure was enhanced by using a molecular sieve and activated carbon. Two molecular sieve tanks are used to absorb water from the biogas. An activated carbon tank (2 meters) was used to eliminate H₂S to < 1 ppm before entering the gas tank. In the form of compressed bio-methane gas (CBG), production was 14,400 m³/day (CH₄ 60-70%), and the amount of CBG was 9,600 m³/day 6.8 tons/day. The raw biogas containing CH₄, CO₂, and O₂ are 68.8%, 29.7% and 0% with H₂S 768 ppm. After the enhancement process, CBG having CH₄, CO₂, O₂ are 89.35%, 10.05 0% and 0.02% with < 0.01 ppm H₂S. Therefore, after the purification process amount of CO₂, H₂S gas was considerably reduced and CH₄ was improved by up to 90% by volume, and then the CBG was compressed to 250 bar tanks to the fuel for cars. Therefore, these results clearly demonstrate that the activated carbon method is feasible to process for the removal of CO₂ and H₂S from biogas in a large-scale plant.

1. Introduction

Biogas is a renewable source of energy that can be used as a substitute for natural gas or liquefied petroleum gas (Souvannasouk et al., 2021). Due to the shortage of fossil energy, biogas is derived from biomass materials, and it is usually used to produce heat and electricity as well as transportation fuels with high efficiency and alternative for natural gas or liquefied petroleum gas (Dussadee et al., 2014; Wannapokin et al., 2017). Therefore, biogas is utilized extensively worldwide as an important renewable energy (Sittisom et al., 2019). Furthermore, previous studies have shown that biogas delivers better environmental benefits than either biodiesel or first generation bioethanol (Junluthin et al., 2021; Patterson et al.,

2011). Biogas was produced through anaerobic digestion (AD) to produce bio-renewable energy. The ability to utilize a wide range of feedstocks, such as biodegradable commercial, industrial, and municipal wastes, represents another potential advantage of fuels produced by AD (Unpaprom et al., 2015; Ramaraj and Dussadee, 2015).

Generally, biogas contains CH₄, CO₂, H₂, H₂S, ammonia, siloxanes and other substances (Ramaraj et al., 2016). Besides CH₄ and CO₂, which are its main components, biogas also contains several trace compounds that must be removed before combustion. For some applications, a certain purity degree of biogas is needed (Unpaprom et al., 2021). And the requirement to upgrade the biogas to biomethane of adequate quality for transport fuel i.e.,

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reducing CO₂ and H₂S content, will significantly improve the quality of biogas (Ramaraj and Dussadee, 2015). The presence of CO₂ and other trace components in biogas could affect engine performance adversely (Dussadee et al., 2016). CO₂ content must be reduced in order to achieve higher heating values and enhance the calorific value of biogas. Furthermore, H₂S is present in biogas produced during the AD of biodegradable substances. It is a major contaminant in biogas produced from the AD of organic materials (Ramaraj et al., 2016). H₂S is produced from the degradation of proteins and other sulfur-containing compounds present in the organic feedstock to the digester.

The concentration of H₂S produced by the digester depends on the feedstock and varies between 0.1 and 2 % (Lastella et al., 2002). It is a highly toxic and malodorous compound, highly corrosive to many types of steel, affects internal combustion engines and considerably shortens the lifetime of the installations for biogas utilization (Li et al., 2007). CO₂ and H₂S can be removed either during the anaerobic digestion process in the digester itself or after the digester, furthermore there are many chemical, physical, and biological methods currently available for removal from biogas (Ramaraj & Dussadee, 2015). The most common methods for biogas enhancement are, air/oxygen dosing to digester slurry, iron sponge, iron oxide pellets, activated carbon, water scrubbing, NaOH scrubbing, biological removal on a filter bed, air stripping and recovery (Sujan et al., 2011).

Since the co-digestion method is widely used to enhance the anaerobic degradations of solid substrates (Unpaprom et al., 2015). In addition, co-digestion is the best choice for high-solid anaerobic digestion (Karapidakis et al., 2010). For co-digestion application, lignocellulosic waste can be digested simultaneously with other materials, such as manure, sludge, and vegetable and fruit waste (Ramaraj and Dussadee, 2015). Napier grass, known as elephant grass (*Pennisetum purpureum*), is a promising bioenergy crop. It is one of the most promising grasses available in tropical and subtropical areas. Hence, the objectives of this research were to produce large amount of biogas yield from swine farms and co-digesting with elephant grass and to investigate CO₂ and H₂S removal processes in practice, furthermore enhancement of CBG production to deliver the transport directly in Thailand.

2. Materials and Methods

2.1 Study site and silage preparation

The compressed bio-methane gas (CBG) plant was in rural area of Mae Taeng District, Chiang Mai Province, Thailand. This investigation is to implement the CBG from swine manure-elephant grass silage co-digestion for vehicles. The methodology is illustrated in Figure 1. Elephant grass (*Pennisetum purpureum*) was collected from the agriculture farm. The farm was cultivated around Mae Tang zone, which is located near the biogas plant. The grass was first cut (cut at an early mature stage, 45 days period). Harvested materials were immediately chopped by the machine into small particles.

Lactic acid bacteria strain (*Lactobacillus plantarum*) was used as silage inoculants. Grass silage was used about 20–23 tons per day, and the silage particle size was 1.0 mm. The grass collecting and silage were prepared.

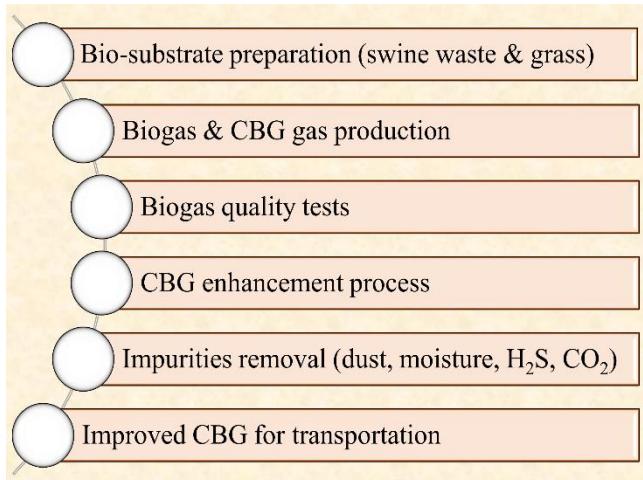


Figure 1 Flow chart of CO₂ and H₂S removal in biogas.

2.2 Material collection, substrates preparations, reactor design and performance

Swine manure and swine farm wastewater were obtained from a swine farm “The sacred pig farm and Sons Farms Ltd” in Mae Taeng district, Chiang Mai, Thailand. The farm has 35,000 pigs and the farm size is about 300 m³. The daily fresh active substrate of 10-12 tons was utilized from the farm. The swine farm substrates (i.e., fresh active substrate, 10-12 tons per day) and grass silage (20–23 tons per day) were pumped through a grinder and then to an equalization tank (150 m³ capacity).

The temperature of the equalization tank was maintained at 40°C, tanks thermal capacity was 600 kWh. This setup related to two anaerobic cultivation tanks (each tank capacity was 20 m³) and a storage/dosing tank. The feed was provided 32,500 kg/24h (as the liquid of 325 m³/24h) to the fermenters.



Figure 2 Industrial scale biogas plant

The use of AD technology in the manure processing industry is predominately centered on wet processes, and more specifically on the utilization of continuously stirred tank reactors (CSTRs), which are the most prevalent and standard reactors (Tsai et al., 2012). We made use of three identical manufacturing units, designated CSTR1, CSTR2, and CSTR3, with each one having a capacity for operating at 1,700 m³. The CSTRs were attached to a stirrer system that could be positioned in one of three distinct ways.

Figure 2 depicts the real views of the reactor assembly from various vantage points. It was permissible to make use of fermenting tanks that maintained a stable temperature and received consistent feed. In addition, the installation of a tank agitator and an agitator control system that was connected to the fermentation tank via PLC (Programmable Logic Controller) allowed for frequent monitoring.

2.3. Biogas enhancements and analytical methods

For the purposes of this investigation, granules of activated carbon of a commercially available medium-grade were utilized. The approximate activated carbon particle size of 2 mm and the measured total surface area and higher heating value (HHV) of about 700 m²/g and 33,600 kJ/kg, respectively. UOP's Separax cellulose acetate membranes were used for CO₂ removal (APHA, 2005). Biogas composition (CH₄, CO₂, H₂, H₂S, and O₂) was measured using an automated gas analyzer according to Brettschneider et al. (2004) and Zhao et al. (2012).

3. Results and discussion

3.1. Biogas production

The biogas plant operating on the principle of a wet anaerobic fermentation process was selected for the composition of input raw material which was determinative for the final biogas quality (Dussadee et al., 2004; Ryckebosch et al., 2011; ASTM, 2010; UNE-EN ISO-6976, 1995). The biogas production takes place during fermentation process in the mesophilic operation (40°C). Daily concentration of individual biogas components is determined. The quantity of CBG from swine manure-elephant grass silage co-digestion biogas produced over a period of 31 days, which is equal to the solid retention time (SRT). During the study period, the biogas ranged from 5,159–15,432 m³, with a mean of 9,605 m³. In addition, biogas production during the test phase is shown Figure 2.

The composition of biogas varies depending on the source (Rasi et al., 2007). Table 1 illustrates its typical composition. Typically, biogas contains 60–65% CH₄, 35–40% CO₂, small amounts of hydrogen sulfide (H₂S), water vapor and traces of other gases. Accordingly, our study results and literature data are listed in Table 1. The evaluations of biogas production show that anaerobic digestion of swine manure and grass silage in CSTRs is feasible. The biogas production was 14,400 m³/day. And CBG production was 6.8 tons/day (i.e., 9,600 m³/day). It contained 60–70% CH₄. The study results showed that the composition of biogas, CH₄, CO₂, O₂ and H₂S contents were found as 68.8%, 29.7%, 0% and 768 ppm, respectively (Rasi et al., 2011).

Table 1 Composition of biogas from different substrates.

Biogas	Composition (%)							Reference
	CH ₄	CO ₂	H ₂ O	H ₂	H ₂ S ppm	NH ₃	O ₂	
Agricultural waste	50–80	30–50	Saturation	0–2	0.70	Trace	0–1	Rasi et al. (2011)
Landfills	47–57	37–41	Saturation	0–5	0.1	Trace	0–1	Karapidakis et al. (2010)
Industrial Waste	50–70	30–50	4–7	0–2	0.8	Trace	0–1	Nges et al. (2012)
Farm biogas plant	55–58	37–38	4–7	<1	<1	Trace	Trace	Rasi et al. (2007)
Sewage digester	61–65	34–38	4–7	Trace	<1	Trace	0–2	Stern et al. (1998)
Biogas plant (without purification)	68.8	29.7	Saturation	<0.01	768	Trace	0.1	This study

3.2. Biogas enhancement and impurities removal

Upgrading to natural gas quality is very much in focus currently as it gives possibilities for alternative applications such as fuel for road vehicles (Ryckebosch et al. 2011). The use of upgraded biogas is considered as one of the most efficient means

of utilizing renewable energy and reducing greenhouse gas emissions (Van Tran et al., 2022). To obtain biomethane of a quality comparable to natural gas with high methane content, it is necessary to further enrichment of biogas. Biogas enhancement can increase the heating value and extend biogas utilization as a renewable fuel (Deng and Hagg, 2010), it would make it possible

to use biogas as an alternative to natural gas. In this study, scrubbing method and CH₄ enrichment process was involved. Nowadays, PSA (Pressure Swing Adsorption) and water scrubbing is the most employed technique for upgrading biogas (Bekkering et al., 2010).

Biogas contains water vapor, and the removal of water vapor is essential as it combines with other contaminants such as hydrogen sulphide or halogenated compounds to produce corrosive acids (Persson, 2003). Gas purification can also be carried out using some form of silica, alumina, activated carbon or silicates, which are also known as molecular sieves (Petersson and Wellinger, 2009). In this study, two molecular sieve tanks are used, and capacity was 3.2 m³ per tank to absorb water from the biogas. By a proper choice of adsorbent, the process can remove CO₂ (Kusworo et al., 2012), H₂S, moisture and other impurities either selectively or simultaneously from biogas (Deublein and Steinhauser, 2008). In addition, the activated carbon tank (2-meter height) was used to reduce H₂S to less than 1 ppm before entering the gas tank. The gas tank size was 200 m³ and the gas pressure was 0.6 bar using Gas compressor package with a 20 bar. It was suitable for CO₂ removal process.

The removal of CO₂ from biogas can be performed by applying adsorption, absorption, cryogenic method, and membrane gas

separation (Brandvoll and Bolland et al., 2004). The first three methods are more traditional compared to the membrane separation, which has some advantages such as low-cost, high-energy efficiency, ease of operation and modular state. Since the first application of cellulose acetate membrane to the CO₂ separation in 1980s (Sujan et al., 2011), various polymeric membrane materials such as cellulose acetate, polyimides, polyamides, polysulfone, polycarbonates, and polyetherimide have been used for the removal of CO₂ from gaseous mixtures. But, cellulose acetate, polyimides and perfluoropolymers have become commercially available for CO₂ removal (APHA, 2005). The UOP Separax cellulose acetate membranes were used for CO₂ removal system was presented in Figure 3. The summary of biogas upgrading performances of different membrane materials comparison is displayed in Table 2.

In the digester biogas containing CH₄, CO₂, O₂ are 68.8%, 29.7% and 0% with H₂S 768 ppm. After enhancement, CBG met the standard of the Department of Energy, having CH₄, CO₂, O₂ are 89.35%, 10.05 0% and 0.02% with < 0.01 ppm H₂S. Consequently, H₂S was removed to below the detection limit and methane content was reached about 90%. Triplicate data of CBG gas analysis are shown in Table 3.

Table 2 Comparison of biogas upgrading performances of different membrane material

Membrane material	Filler	Effect of fillers on CO ₂ /CH ₄ separation	Reference
Acrylonitrile-butadiene-styrene	Activated carbon	Improved	Anson et al. (2004)
Aluminosilicate gel	T-type zeolite	Improved	Mirfendereski et al. (2008)
Aromatic poly (amide-imide)	TiO ₂	Improved	Hu et al. (1997)
Matrimid	MgO	Improved	Hosseini et al. (2007)
Matrimid	Carbon aerogel	Improved	Zhang et al. (2008)
Matrimid® 5218	TiO ₂	Improved	Moghadam et al. (2011)
Matrimid® 5218	Carbon molecular sieve	Improved	Vu et al. (2003)
Polyimide	Silica	Improved	(Suzuki & Yamada, 2005)
Polyvinyl acetate	Zeolite 4A	Improved	Zornoza et al. (2011)
Polysulfone	Metal organic frameworks	Improved	Zornoza et al. (2011)
Polyethersulfone	Zeolite NaA	Improved	Cakal et al. (2012)
Polyethersulfone	2-Hydroxy 5-methyl aniline	Improved	Cakal et al. (2012)
Polyethersulfone	Carbon nanotube	Improved	
Rubber	Silicalite-1	Improved	Duval et al. (1993)
	13X and KY	Improved	
	Carbon molecular sieve	Not improved	
Ultem® 1000	Carbon molecular sieve	Improved	Vu et al. (2003)
UOP Separax cellulose acetate	Carbon molecular sieve	Improved	This study

Table 3 Composition of gas through the gas quality improvement

Experiment	Gas before the gas composition through quality improvement			
	CH ₄ (%Vol.)	CO ₂ (%Vol.)	O ₂ (%Vol.)	H ₂ S (mg/m ³)
1	91.61	8.31	0.01	<0.046
2	85.39	14.28	0.04	<0.046
3	85.46	14.34	0.01	<0.046
Average	87.49	12.31	0.02	<0.046

**Figure 3** UOP Separax cellulose acetate membranes CO₂ removal system

By removing carbon dioxide, moisture, hydrogen sulfide and other impurities, biogas can be upgraded to biomethane, a product equivalent to natural gas. Biomethane can be used as compressed natural gas (CNG) in natural gas vehicles. Compressed natural gas (CNG) is a form of natural gas storage that is stored at high pressure of around 200 bar. Compressed biomethane gas (CBG), is equivalent to compressed natural gas (CNG). In the form of which

could be biogas upgraded to biomethane and subsequently used as a transport fuel in a CNG vehicle. Compressed biomethane is almost identical to compressed natural gas, which is currently used as a transport fuel in many countries worldwide such as Sweden and Austria. The system performance and biogas enhancement data are given in Table 4.

Table 4 Performance test system, improve quality biogas.

Parameter	Analysis result			The standards of the Department of Energy Business
	Before the system improve gas quality	Back through the system, improve gas quality	Efficacy (%)	
CH ₄ (% vol)	61.89	87.49		≥ 65

CO ₂ (% vol)	34.12	12.31	63.92	≤ 18
O ₂ (% vol)	0.10	0.02	80	≤ 1
H ₂ S (mg/m ³)	1,578	<0.046	100	≤ 23

Furthermore, these countries are also using compressed biomethane as a standard transport fuel. Production of CBG was used as automotive fuel for NGV substitution. This study examined the use of CBG for cars in Mae Taeng District. The upgraded biogas will be used for vehicles in rural areas. CBG production units are installed on pig farms in Mae Taeng District, Chiang Mai Province, Thailand.

4. Conclusions

Results obtained in this work demonstrate that it is possible to achieve the complete removal of H₂S and CO₂ from biogas using activated carbon for an absorption process that operates at ambient temperature and UOP Separax cellulose acetate membranes. This study shows that it is possible to develop a procedure for removing H₂S from compressed biogas by activated carbon, separately in a filter bed. Furthermore, the results presented show that the almost selective increased rate of H₂S removal is not the only advantage of this process of chemical absorption. The main advantage is the transformation of H₂S into S, thereby eliminating the pollution potential of H₂S.

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