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Study of on-site upgraded livestock biogas production and carbon emission reduction by substituting coals for thermal power generation

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ABSTRACT

The objective of this project is to integrate a farm-scale bio-desulfurization facility with a novel biogas hollow fibre adsorption module for biogas desulfurization and bio-natural gas production. In this study, the desulfurization experimental results showed that the bio-desulfurization system can remove 96.7±6% of H_2S from the biogas after an approximately two-month enrichment period. The average CH_4, N_2, and CO_2 concentrations in raw biogas were 63.4, 15.2, and 21.1%, respectively. As for biogas upgrading experiments, the inlet biogas flow rates were applied from 5 to 20 L/min. The removal efficiency of CO_2 under all biogas flow rates was 100%. Meanwhile, methane was promoted from 60% to nearly 94% (i.e., 57% increase in methane concentration). The replacement of anthracite and coking coal by upgraded biogas might reduce 44.4% and 42.5% of CO_2 equivalent, respectively. The achievement of this project pursues the mitigation of carbon dioxide emissions by using upgraded pig biogas which can be enlarged and extended to all decentralized pig farms worldwide.

Introduction

World meat production reached 337 million tonnes in 2020, up 45 percent, or 104 million tonnes compared with 2000 (FAO, 2022). Pig meat represented 33 percent of the total in 2020, compared to 39 percent in 2000 (FAO, 2022). African swine fever started affecting Asian countries in late 2018 and continued in 2019 and 2020. It resulted in an 11 million tonnes decrease in world pig meat production between 2018 and 2019 but kept constraining production in 2020 (FAO, 2022). Pig slurry or piggery wastewater can be anaerobically treated to produce biogas as a potential sustainable cleaner energy.

Pig farming is the dominant livestock industry in terms of production value totaling around US$5.6 billion, i.e., about 33% of agricultural production value (US$16.8 billion) in 2021 (COA, 2021). Moreover, pig farming production value (USD 2.4 billion) is about 43% of livestock production value (USD 5.6 billion) in 2020 (COA, 2021). In 2022, Taiwan had 6,133 pig farms with a total of 5,375,109 pigs (COA, 2022). However, Taiwan had only 1,540 pig farms (25% of the total farms) with more than 1000 pigs each. These farms constitute 71.5% (3,844,139 pigs) of the total number of pigs. The organic waste produced from livestock farms including animal manure, sludge, wastewater, and other organic wastes can be recycled as a re-

newable energy source by anaerobic digestion (AD) to avoid polluting the environment (Su, 2020). The collection of biogas from pig farms is used for power generation or direct combustion (Su and Chen, 2015). Anaerobic co-digestion of animal manure and crops provides a combined production of renewable energy and organic fertilizer on organic farms and has been suggested as an option to improve the operation model of organic agriculture. The analytical results of anaerobic digestion and biogas production on a 1000-ha model farm with combined dairy and cash crop production in Denmark were presented (Pugesgaard *et al.*, 2013). The effects on crop rotation as well as greenhouse gas (GHG) emissions were assessed for four scenarios of biogas production with animal manure. The authors concluded that the production of biogas on the experimental farms holds the possibility for the farms to accomplish a positive energy balance providing self-support with organic fertilizer nitrogen and decreasing GHG emissions.

In Taiwan, however, biogas is mainly produced from livestock farms livestock biogas with livestock wastewater, i.e. animal manure mixed with cleaning water, after solid/liquid separation. Thus, the amount of biogas was less than that without solid/liquid separation and the contents of biogas from piggery wastewater were about 60-70 % CH_4, 30 %CO_2, and a little H_2S (Su *et al.*, 2003; Su and Chen, 2018). The H_2S in biogas can corrode engines and gas pipelines (McCarthy, 1998). On the other hand, the high content of CO_2 in biogas reduces its heating value and restricts its applications (Abatzoglou and Boivin, 2009). These gases, CO_2 as well as H_2S, can decrease biogas combustion efficiency and cause corrosion problems, respectively.

For biogas desulfurization purposes, a novel farm-scale biogas bio-desulfurization system (BBS) was developed and applied for on-site livestock biogas desulfurization by the research team of Animal Technology Institute Taiwan (ATIT) (Su *et al.*, 2008) and National Taiwan University (NTU) (Su *et al.*, 2013, 2014). Additionally, a biogas photocatalytic desulfurization reactor (PDR) was also successfully developed for small-scale biogas desulfurization purposes (Su and Hong, 2020) as well as in coordination with pilot-scale hollow fibre adsorption module (HFAM) for CO_2 removal by the research team of NTU (Su and Chung, 2021).

For biogas upgrading purposes, there are some applications for upgrading desulfurized biogas, including pressurized water scrubbing, amine swing absorption, pressure swing adsorption, temperature swing adsorption, cryogenic separation, and membrane technologies. The main operations available on the market are absorption, adsorption, cryogenic separation, and membrane separation (Chen *et al.*, 2015). Absorption can be classified into physical and chemical methods. High-pressure water scrubbing and organic physical scrubbing are physical absorptions (Cozm *et al.*, 2013). Physical absorptions have advantages such as simplicity, high methane purity, less methane loss, and low operating and maintenance costs (0.26 USD/m^3 biogas). However, they require large amounts of water, high energy demand, chances of biological contamination, and external heat (Adnan *et al.*, 2019). Chemical absorptions, such as amine scrubbing and inorganic solvent scrubbing, can dissolve more CO_2 per unit volume and have a faster processor, but they require high energy to produce steam (0.44 USD/m^3 biogas), pre-treatment, and difficulties in handling solvents (Adnan *et al.*, 2019). Adsorption operations can be characterized as pressure swing adsorption, vacuum swing adsorption, temperature swing adsorption, and electrical swing adsorption. Adsorption operations have advantages such as high gas quality, low methane losses, dry process, low energy demand (0.26 USD/m^3 biogas), and no chemical use. However, they are complex, require pre-treatment, and need multiple streams to increase biogas quality (Adnan *et al.*, 2019). Cryogenic operation is a distillation process operated under very low temperatures (close to -170°C) and high pressure (around 80 bar). This process allows introducing of high flow rates of raw biogas and reaching high CH_4 concentrations in the range of 90−99 %. Cryogenic separation produces high-quality biogas and liquid bio-methane with low additional energy but has high investment and operational costs, requires pre-treatment and high energy for cooling, and is still under development (Adnan *et al.*, 2019). For membrane technology, it is based on gas di□usion and dissolution into polymer materials. Membrane separation has low energy consumption, low cost, and a simple process but has low membrane selectivity, requires pre-treatment, and results in low methane purity. (Adnan *et al.*, 2019; Chen *et al.*, 2015). To promote CH_4 concentration in the desulfurized biogas, a hollow fibre CO_2 adsorption system was introduced and applied for biogas upgrading. (Vogler *et al.*, 2013; Falbo *et al.*, 2014; Ibrahim *et al.*, 2018; Žák *et al.*, 2018 Tantikhajorngosol *et al.*, 2019; Su and Chung, 2021).

A feed-in tariff (FIT) is a policy tool designed to promote investment in renewable energy sources such as photovoltaics, wind, waste, biomass, geothermal, marine, or hydro. Sustainable investing directs investment capital to companies that seek to fight climate change and environmental devastation while promoting corporate responsibility. A FIT can be applied to international market applications including the energy market, energy infrastructure, and technological conditions of each country. However, FITs usually expire after a limited period, because of the irregular electricity supply of renewable electricity sources (RESs), market alterations, and insufficient flexibility options, limited participation in wholesale electricity markets is a viable business model for RES no longer receiving FITs (Rövekamp *et al.*, 2021).

The objective was to study the feasibility of combining a farm-scale bio-desulphurization facility with a hollow fibre CO_2 adsorption module (HFAM) for biogas upgrading. The acceptable H_2S in the biogas for livestock biogas applications should be lower enough and not harm facilities. The upgraded biogas promoted in situ livestock biogas applications such as power generation or fuel for heat preservation.

Materials and Methods

Pig farm for this study

The selected commercial pig farm, the I-Yang Pig Farm raises about 1,500 pigs, and was in I-Lan County, Taiwan (https://www.facebook.com/iyangranch/ - accessed on 03 January 2022). The daily wastewater was about 48 m^3/day. Analytical data of the wastewater samples showed that the suspended solids (SS), chemical oxygen demand (COD), and biochemical oxygen demand (BOD) of the raw wastewater after solid/liquid separation were 176±76, 2262±450, and 846±279 mg/L, respectively. The biogas produced from the wastewater treatment facility of this pig farm was used for this study.

Farm-scale biogas bio-desulfurizing system

The farm-scale biogas bio-desulfurizing system (BBS) was installed on the I-Yang Pig Farm. All processes of biogas desulfurization and upgrading are shown in Figure 1. The automatic operation design of BBS was modified from the design of a previous

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study (Su *et al.*, 2013). The modified BBS was constituted of a bio-filter (1 m outer diameter × 6 m height) filled with Raschig rings (i.e. hollow spherical polypropylene balls) (Sheng-Fa Plastics, Inc., Taiwan) and lightweight expanded clay aggregates (LECA). The condensed water from the bottom of the BBS was collected and analysed by ion chromatography (IC) as well as pH/conductivity check periodically. After the water quality of condensed water was stably acidic (e.g. pH < 2), the gas samples were collected from the inlet and outlet of BBS for gas chromatography (GC) analysis. H_2S in biogas was measured by a portable multi-gas detector or H_2S detector tubes for calculating the H_2S removal by the BBS.

Dehydration and filtration reactor (DFR) and activated carbon reactor (ACR)

The DFR was constituted of a dehydration reactor and three filtration reactors (115 cm height × 30 cm inner diameter, total volume = 81 L) (Figure 2). The dehydration reactor was packed with a mixture of Raschig rings and LECA. The ACR was the same container as DFR but filled with coconut shell-activated carbon pellets (diameter = 5 mm) and Raschig rings. The DFR and ACR were applied to remove excessive moisture and volatile organic compounds from the desulfurized biogas. The operation pressure was under ambient pressure.

Farm-scale hollow fibre CO_2 adsorption module (HFAM) for biogas upgrading

The farm-scale hollow fibre CO_2 adsorption module (HFAM) was installed followed by the farm-scale BBS in series. The HFAM (AuraMat-HFD CO2-CH4-15L-VC, Aura Material Inc., Hsinchu, Taiwan) (100 cm width × 85 cm length × 135 cm height, power voltage = 220 V 60/Hz) was constituted of two sets, Set #A and B, of hollow fibre CO_2 adsorption cartridges (60 cm length × 3" outer diameter) (AuraMat-HTP-CO2-A3, Aura Material Inc., Hsinchu, Taiwan) in parallel. The size of all biogas tubing, including biogas inlet, biogas purging, and biogas outlet tubing, was 1/2" tubing. Each adsorption cartridge set constitutes eight hollow fibre CO_2 adsorption cartridges in series. Two gas sampling ports were installed at the biogas inlet and outlet tubing for periodical biogas sampling. Desulfurized biogas was introduced into the module through two filtration cartridges, AuraMat-TP-HC-A-B and AuraMat-TP-HC-Oil-S (Aura Material Inc., Hsinchu, Taiwan) by an explosion-proof vacuum pump (flow rate = 15~26 L/min, No. N 026, KNF Neuberger, Inc., Trenton, NJ, USA) inside the module. The operation pressure of the CO_2 adsorption cartridge was <1~2 kg/cm^2.

The sequencing of adsorption and regeneration processes for the HFAM was shown in Figure 3 a,b. Figure 3a represents the adsorption process by Set A and the regeneration process by Set B simultaneously. Moreover, Figure 3b represents the adsorption process by Set B and the regeneration process by Set A simultaneously. Therefore, the operation process of Set A was as follows in sequence: desulfurized biogas from the storage bags, filtration cartridge (AuraMat-TP-HC-A-B), hollow fibre adsorption cartridges (AuraMat-HTP-CO2-A3), and then discharging upgraded biogas (Figure 3a, solid black line).

When hollow fibre CO_2 adsorption cartridges of Set A were saturated, the inlet biogas was automatically shifted to set B following the operation process for biogas upgrading (Figure. 3a, solid black line). Simultaneously, the hollow fibre CO_2 adsorption cartridges of Set A followed the regeneration process and were automatically regenerated. Therefore, the regeneration process of the HFAM was as follows in sequence: purged air, filtration cartridge (AuraMat-TP-HC-Oil-S), hollow fibre adsorption cartridges (Au-

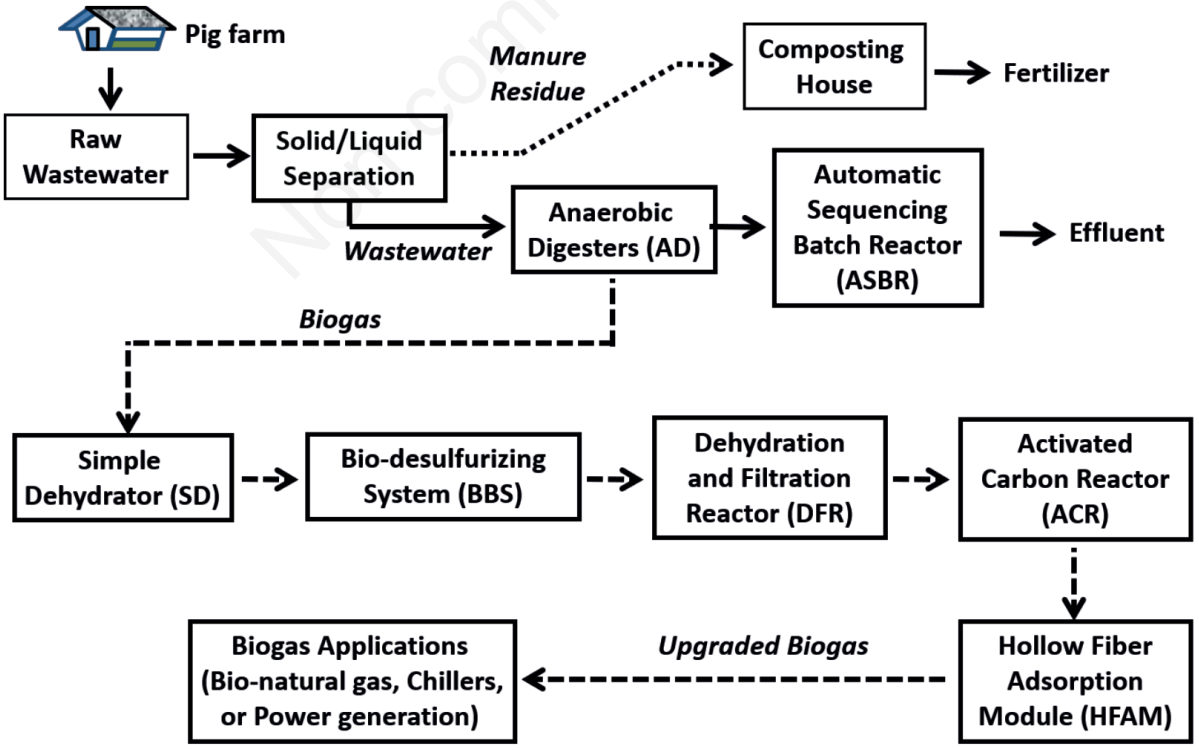

Figure 1. Flowsheet of all processes for biogas production, desulphurization, and upgrading.

raMat-HTP-CO2-A3), vacuum pump, and then discharging gas (Figure 3b, blue dashed line). Similarly, once the hollow fibre CO_2 adsorption cartridges of Set B are saturated, the operation process will automatically switch to the regeneration process (Figure 3a, blue dashed line). Thus, Sets A and B were operated alternately.

Biogas upgrading using hollow fibre CO_2 adsorption module with automatic regeneration

The HFAM had to be warmed up for at least 2.5 h and then another 2.5 h for filling up biogas inside the cartridges of the CO_2 adsorption module before starting any time-course experiments. Desulphurized biogas was introduced into the biogas filtration cartridge (AuraMat-TP-HC-A-B, Aura Material Inc., Hsinchu, Taiwan) and then through eight independent hollow fibre CO_2 adsorption cartridges (AuraMat- HTP-CO2-A3, Aura Material Inc., Hsinchu, Taiwan) of the Set A or B through an explosion-proof vacuum pump inside the adsorption module for time-course experiments of CO_2 removal from the biogas. Biogas samples were taken at 10 min intervals in an hour from the inlet and the outlet of the CO_2 adsorption cartridge module under the outlet biogas flow rates of 5, 10, 15, and 20 L/min. The hollow fibre CO_2 adsorption cartridges of Sets A and B have operated alternatively and regenerated automatically based on the signals on the control panel. Biogas samples were taken at 10 min intervals in an hour from the inlet and outlet of the adsorption cartridge module under various outlet biogas flow rates. The contents of all biogas samples were determined by using GC with a thermal conductivity detector (GC/TCD) (Su and Chung, 2021).

Analysis

Determination of pH, EC, and ions in liquid samples

Electrical conductivity (EC) and the pH of water samples were determined by a pH/conductivity meter (ExStik EC500, EXTECH Instrument, FLIR Commercial Systems, Goleta, CA, USA). Ion concentrations of water samples were determined by anion chromatography (IC) (Metrohm, 883 Basic IC Plus, Switzerland) equipped with an auto-sampler (863 Compact Autosampler). Anions were separated by Metrosep A Supp 5 (4 mm × 150 mm) column, and included SO_4^{2-}, PO_4^{3-}, NO_3^-, NO_2^- and Cl^-. Samples were diluted and filtered through a 0.2 mm filter before injecting them into ion chromatography.

H_2S determination in biogas samples

A portable multi-gas detector (ISC MX series, Industrial Scientific Co., PA, USA) was used to measure the concentration of H_2S on-site of the biogas samples from both inlet and outlet of the BBS. When the concentration of H_2S was over the detection limit of the portable multi-gas detector (sensor: H_2S: 0-697 mg/L, SO_2: 0-393 mg/L, O_2: 0-0.30 (% vol.), NH_3: 0-348 mg/L), a gas sampling pump (GV-100C gas sampling pump; Gastec Co., Japan) with H_2S detector tubes (H_2S = 14-5575 mg/L) (Gastec Co., Kanagawa, Japan) was applied for H_2S detection.

CH_4, CO_2 and N_2 determination in biogas samples

Tedlar® bags (SKC, PA, USA) with a single polypropylene fitting were used for collecting biogas samples from the inlets and outlets. The fitting contained a Teflon® syringe port-lined septum and a hose connection, which functioned as a shut-off valve for incoming and outgoing gas. Meanwhile, a 500 mL gas collector (GL Sciences Inc., Tokyo, Japan) was used to withdraw gas samples from the inlets and outlets of the PDRs. Biogas samples were analysed for their composition by GC (Master GC, DANI Instruments, Marlborough, MA, USA), which was equipped with a TCD and Carboxen 1010 PLOT capillary column (30 m× 0.53 mm× 0.25 µm film thickness; Supelco Analytical of Sigma-Aldrich Co., PA, USA) (Su and Chung, 2021). Calibration curves of CH_4, CO_2, and nitrogen gas were obtained by the external standard method, and the calibration curve's correlation coefficient is >0.995.

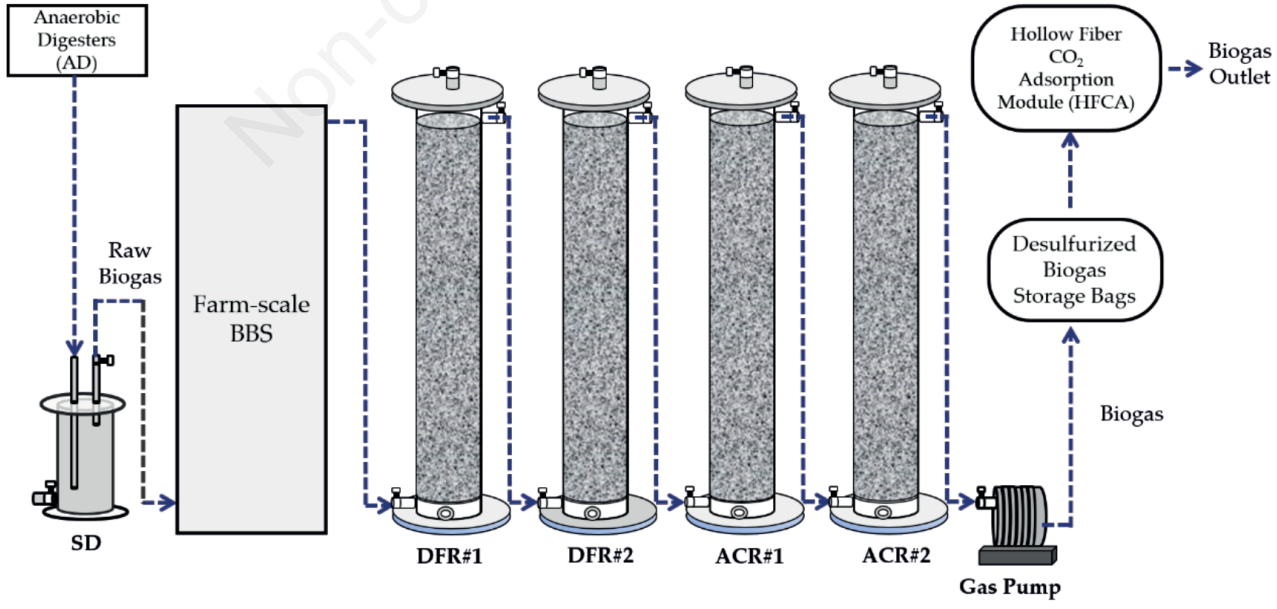

Figure 2. Sketch and flowchart of the biogas bio-desulfurization system (BBS) in coordination with a biogas dehydration and filtration reactor (DFR) and hollow fibre CO_2 adsorption (HFCA) module.

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Estimation of carbon emission reduction

Estimation of carbon emission was carried out by applying clean development methodology and emission factors of stationary combustion (IPCC, 2006b). The baseline scenario and project scenario assumed coking coal or anthracite (100 tons/year) and upgraded biogas as the fuels for thermal power generation, respectively. The net caloric values (NCV) of biogas (refer to as slurry gas), anthracite, and coking coal as well as the global warming potential (GWP) of CO_2, CH_4, and N_2O were used for both baseline and project scenario carbon emission (Table 1). Moreover, the emission factors (EFs) of CO_2, CH_4, and N_2O for

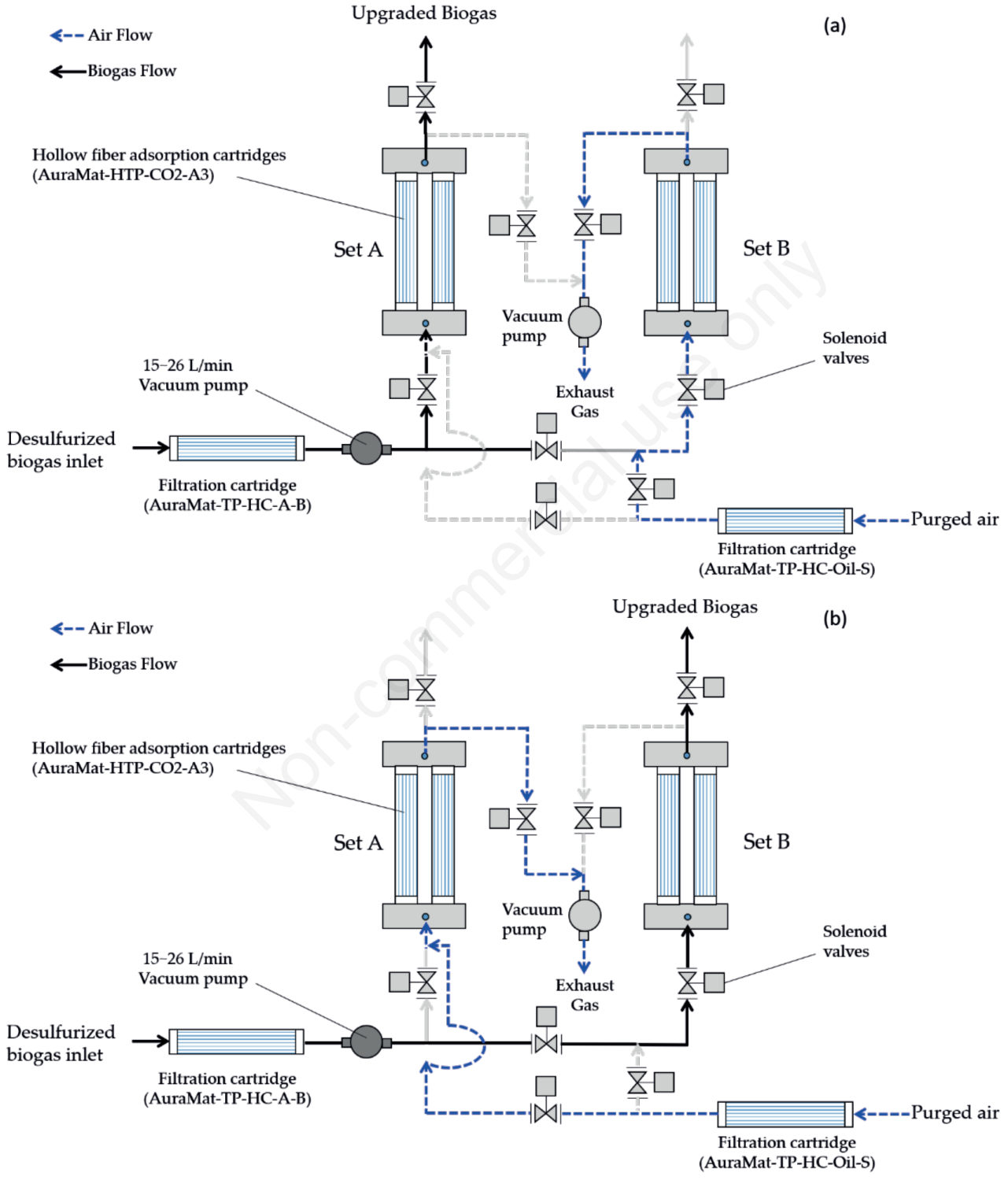

Figure 3. Flow chart of the hollow fibre CO_2 adsorption system including operation process (in solid lines) and regeneration process (in dashed lines). Where Set A is under operation process (a) or Set B is under operation process (b).

stationary combustion of biogas (as slurry gas), anthracite, and coking coal are used for calculation (Table 2). The carbon emission of the baseline scenario (t CO_2e/yr) is calculated based on EFs of coking coal or anthracite. Moreover, the carbon emission of the project scenario (t CO_2e/yr) is calculated by replacing cocking coal or anthracite with biogas (slurry gas) and based on EFs of slurry gas as follows:

Baseline scenario = 100 tons/yr × NCV (coking coal or anthracite) × EF (coking coal or anthracite) × GWP_{100}

Project scenario = 100 tons/yr × NCV (coking coal or anthracite) × EF (slurry gas)× GWP_{100}

Statistical analysis

Experimental data of water quality at different operation days and gas concentrations at different flow rate levels were analysed with one-way analysis of variance (ANOVA) by Origin 2020b software. When ANOVA identified a significant effect, Tukey's test was applied for multiple comparison tests. The Student's *t*-test was applied to the data of the bio-desulfurization test and gas concentrations between the inlet and outlet of the CO_2 adsorption module. The significance level is 0.05.

Results and Discussion

The initiation of BBS and biogas desulfurization

The water quality indexes of dropping water, i.e., condensed liquid, from BBS at different operation days were significantly different over time except for nitrite and chloride (Table 3). The dropping water samples were from the oxidation of H_2S in biogas into the condensed liquid. Thus, the pH value of dropping water, 0.78±0.16 - 4.85±0.69, decreased over operation time resulting from increased SO_4^{2-} concentrations ($p<0.05$). Results showed that SO_4^{2-}, PO_4^{3-}, NO_3^-, NO_2^-, and Cl^- in dropping water over operation time were 1,873±197 - 14,462±1038 mg/L ($p<0.05$), 286±58 - 511±78 mg/L ($p<0.05$), 27±25 - 102±30 mg/L ($p<0.05$), 88±62 - 147±12 mg/L, and 57±39 - 106±6 mg/L ($p<0.05$), respectively. The EC of dropping water over operation time was 5.56±1.81 to over 400 mS/cm (<0.05) (Table 3). Thus, increased EC in the dropping water samples is closely related to the increase of both SO_4^{2-} and PO_4^{3-} concentrations.

The biogas samples from the inlet and outlet of BBS were collected and analysed after 60 operation days (Table 4). The

Table 1. Net caloric value (NCV) for fuels and global warming potential (GWP) (IPCC, 2006a, 2007).

Fuels	NCV (TJ/Gg)		
Gasoline	44.3		
Slurry gas	50.4		
Anthracite	26.7		
Coking coal	28.2		
Global warming potential (GWP100)			
GHGs	CO_2	CH_4	N_2O
CO_2e	1	25	298

Table 2. Default emission factors for stationary combustion of fuels (IPCC, 2006b).

Fuels	Emission factors for stationary combustion (kg of greenhouse gas per TJ on an NCV basis)		
	kg-CO_2/TJ	kg-CH_4/TJ	kg-N_2O/TJ
Gasoline	69,300	3	0.6
Anthracite	98,300	1	1.5
Coking coal	94,600	1	1.5
Slurry gas	54,600	1*	0.1

NCV, net caloric value.

Table 3. Continuous data of pH, electrical conductivity, and anions of the dropping water samples from a farm-scale biogas bio-desulfurization system (n=20).

Operation time (days)	pH	EC (mS/cm)	SO_4^{2-} (mg/L)	PO_4^{3-} (mg/L)	NO_3^- (mg/L)	NO_2^- (mg/L)	Cl^- (mg/L)
2	4.85±0.69	5.56±1.81	1873±197	334±57	102±30	123±86	68 57
5	2.66±0.15	7.76±0.18	2819±164	286±58	62±41	88±62	106±6
7	0.78±0.16	>400	14462±1038	511±78	27±25	147±12	57± 9
p-value	<0.05	<0.05	<0.05	<0.05	<0.05	NS	NS

EC, electrical conductivity; NS, not significant.

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un-desulfurized biogas contains 1967 ± 512 mg/L H_2S. After passing through BBS, the H_2S of biogas was removed by 96.7 ± 6%. On the other hand, the desulfurized biogas only remained at 67 ± 121 mg/L H_2S (Table 4). In addition, the concentrations of N_2, CO_2, and CH_4 showed no significant difference between the biogas before and after desulfurization by BBS.

The results showed that the pH value of dropping water from BBS decreased and its EC and SO_4^{2-} increased as the operation time went on. The H_2S removal from biogas by the BBS is followed by a series of oxidation-reduction reactions ($H_2S \rightarrow S^0 \rightarrow SO_4^{2-}$). According to a previous study, when dropping water had a pH < 2, the sulfur-oxidizing bacteria formed stable biofilms (Su *et al.*, 2013). Thus, in this study, the dropping water from BBS became acid and contained high SO_4^{2-} indicating the system removed H_2S from biogas successfully. As expected, the further results of H_2S removal revealed that the BBS worked very well in removing H_2S from untreated biogas. This study demonstrated that the pH value of dropping water is a good indicator of BBS initiation.

The flow rate experiment of hollow fibre CO_2 adsorption for biogas upgrading

The gas composition of biogas samples from the inlet and outlet of the HFAM is presented in Table 5. Applied 5, 10, 15, and 20 L/min inlet flow rates to the module, the inlet CO_2 in desulfurized biogas was 34.3±2.6 - 34.7±1.5% and the removal efficiency of CO_2 was 100%, i.e. the outlet CO_2 in upgraded biogas was 0.0±0.0 % (Table 5). However, the inlet CH_4 in desulfurized biogas was 60.0±0.3 - 61.2 1.3% and the outlet CH_4 in upgraded biogas was 92.9±2.1 - 93.8±0.6%. As for N_2 content, the inlet desulfurized biogas was 4.5±1.3 - 5.5±0.3% and the outlet upgraded biogas was 6.2±0.6 - 7.1±2.1% (Table 5). The results implied that the HFAM module can only remove CO_2 efficiently and remained CH_4 and N_2 in the outlet biogas. Thus, changes in CH_4 concentrations between the inlet and outlet biogas samples after the upgrading process at any inlet flow rates ($p<0.05$).

The experimental results indicated that the HFAM was capable to remove CO_2 from the desulfurized biogas and promoting CH_4 concentrations within 5–20 L/min inlet flow rates. The CO_2 removal tendency of the HFAM for desulfurized biogas was the same as that of our previous pilot-scaled study (Su and Chung, 2021). The results showed that the methane purity of the previous study and this study was 86% and 93.6%, respectively (Su and Chung, 2021). Also, the CO_2 removal efficiency of the previous study and this study was 97.0% and 100%, respectively. This study with higher inlet biogas flow, i.e. higher biogas treatment volume, but consumed less electric energy (3.4 kWh/Nm^3 CH_4) than our previous study (18.7 kWh/Nm3 CH_4) (Su and Chung, 2021) (Table 6). It implies that the higher the biogas treatment volume, the lower the operation cost of biogas upgrading. Moreover, a comparison of other hollow fibre membrane technique studies showed that the product CH_4 purity of this study was lower (93.6 %) than that of Žák *et al.* (2018). However, our study performed better CO_2 removal (100 %) and operated under ambient temperature and lower pressure with less energy consumption. The low CH_4 purity of the product might be owing to higher N2 concentration in raw biogas. This limited the CH_4 purity in product gas after the up-

Table 4. The difference in biogas composition after bio-desulfurization (n=12).

	H_2S (mg/L)	Removal (%)	N_2 (%)	CH_4 (%)	CO_2 (%)
Raw biogas	1967±512	-	14.9±1.2	63.3±1.9	21.9±0.9
Bio-desulfurized biogas	67±121	96.7±6	15.2±1.2	63.7±2.7	21.1±1.6
p-value	<0.05	-	NS	NS	NS

Table 5. Carbon dioxide removal of the desulfurized biogas by hollow fibre (CO2) adsorption module (n=12).

Inlet biogas (L/min)	CH_4 (%) Inlet	CH_4 (%) Outlet	CH_4 (%) Difference	CH_4 (%) p-value	N_2 (%) Inlet	N_2 (%) Outlet	N_2 (%) Difference	CO_2 (%) Inlet	CO_2 (%) Outlet	CO_2 (%) Removal
5	60.0±0.3	93.7±1.1	+56.1	<0.05	5.5±0.3	6.3±1.1	+13.9	34.5±0.2	0.0±0.0	100
10	60.8±1.7	93.2±0.7	+53.4	<0.05	4.5±0.8	6.8±0.7	+50.0	34.7±1.3	0.0±0.0	100
15	61.2±1.3	92.9±2.1	+51.9	<0.05	4.5±1.3	7.1±2.1	+59.0	34.3±2.6	0.0±0.0	100
20	60.3±2.2	93.8±0.6	+55.7	<0.05	5.0±0.7	6.2 0.6	+24.2	34.7±1.5	0.0±0.0	100

Table 6. The comparison of biogas upgrading indicators with the previous study.

	Scale	CH_4 purity (% vol.)	CO_2 removal (% vol.)	Operating conditions	Inlet biogas flow (L/min)	Energy consumption (kWh/Nm^3 CH_4)
This work	Farm	93.6	100	2 bar, ambient temperature	20	3.4
Su and Chung (2021)	Pilot	86	97.0	Ambient pressure and temperature	1	18.7

grading process. Therefore, additional N_2 adsorption by a specific adsorption module should be considered for promoting higher CH_4 concentrations in upgraded biogas.

Evaluation of upgraded biogas for power substitution

The revenue for electric power generation in different scenarios, 60%, 90%, and 95% methane in biogas, with either 30-kW or 65-kW generators, was shown in Table 7. Based on the 2023 feed-in tariff (FIT) for the biogas power generation from anaerobic digesters in Taiwan (0.24 USD/kWh) (Taiwan Ministry of Economic Affairs, 2023) and the operation time of the power generator (350 d/yr), the revenue using a 30-kW generator was 30.2, 45.4, and 47.9 thousand USD/yr, respectively. On the other hand, the revenue using a 65-kW generator was 65.5, 98.3, and 103.7 thousand USD/yr, respectively (Table 7).

The fuel value of bio-neutral gas equal to liquefied petroleum gas (LPG) was calculated below: the maximum flow rate (20 L/min) was used to gain daily methane yields of 21.7 m3/d (Table 8). Then, the volume of LPG (20 kg/barrel) is 9,391 L per barrel, where the density of LPG (butane) is 0.575 kg/L at 15oC (Afrox, 2022) and 1 L-LPG can be expanded to produce 270 L-gas based on the data of Elgas limited (2021). The daily methane yields were converted into LPG barrel yields, which is 2.31 barrels/d. With further calculation, it is 843 barrels/yr (Table 8).

Estimation of installation and operation costs for biogas upgrading

The initial installation cost (78,284 USD) included re-generable (HFAM) (76,321 USD per set), dehydration and filtration reactor (DFR), and activated carbon reactor (ACR) (1,963 USD/set) (Table 9). Because several types of biogas desulfurization systems were adopted by different livestock farms with various installation costs, the installation cost of the farm-scale biogas bio-desulfurization system (BBS) was not included and calculated.

The annual operation cost (total cost = 4357 USD/yr) in-

Table 7. Estimation of the revenue for power production under different methane concentrations in biogas with either a 30-kW or 65-kW generator.

Methane in biogas (%)	Generator capacity (kW)	Operation time (h/d)	Daily power production* (kWh/d)	Efficiency (%)	Revenue for power generation# (thousand USD/yr)
60	30	20	360	100	30.2
90			540	150	45.4
95			570	158	47.9
60	65	20	780	100	65.5
90			1,170	150	98.3
95			1,235	158	103.7

*Daily power production, methane in biogas × generator capacity × operation time; #revenue for power generation = daily power production × operation time (350 d/yr) × feed-in tariff (0.24 USD/kWh in Taiwan in 2023).

Table 8. Estimation of the fuel values using upgrade biogas instead liquid petroleum gas (LPG).

Item	Value	Calculations	Item	Value	Calculations
Daily methane yields	21.7 m^3/d	20 L/min × 60 min/h × 24 h/d × 80% productivity × 94% purity = 21.7 m^3/d	The volume of liquefied petroleum gas (LPG) (20 kg/barrel)	9,391 L/barrel	20 kg/barrel ÷ 0.575 kg/L × 270 L-gas/ L-LPG = 9391 L/barrel
Converting daily methane yields to LPG yields	2.31 barrels/d	21.7 m^3/d ÷ 9.4 m^3/ barrel = 2.31 barrels/d	Converting yearly methane yields to LPG yields	843 barrels/yr	2.31 barrels/d × 365 d/yr = 843 barrels/yr

Table 9. Estimation of total costs for a farm-scale biogas upgrading system.

Installation cost		Operation cost	
Items	Price (USD)	Items	Price (USD/yr)
Installation of BBS	(35,714)	Electricity cost of HFAM	2,571
Installation of HFAM	76,321	Replacement of the packing materials (DFR and ACR)	357
Installation of DFR and ACR	1,963	Replacement of the adsorption and filtration cartridges (HFAM)	1,429
Total	78,284		4,357

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cluded the electricity cost of the HFAM, replacement of the adsorption, and filtration cartridges of HFAM (357 and 1,429 USD /yr, respectively). Thus, the daily operation cost not including installation cost is 12.1 USD/d. The upgraded biogas cost is 0.0004 USD/L (Table 9).

A previous study using a pilot-scaled and integrated photocatalytic biogas desulfurizer and hollow fibre CO_2 adsorption module for the biogas flow rate of 1 L/min was successfully developed and tested (Su and Chung, 2021). The initial installation and operation cost of the pilot-scaled HFAM is 21,970 USD and 2,028 USD/yr, respectively (unpublished data). Moreover, at another on-planning full-scale centralized demonstration site for livestock biogas upgrading for the biogas flow rate up to 2000 L/min, the initial installation cost was about 183,333 USD (unpublished data provided by Aura Material Inc., Hsinchu, Taiwan). In other words, the higher the biogas flow rates for upgrading, the less the marginal installation and operation cost of the HFAM facility.

Estimation of carbon emission reduction

Assuming the use of coking coal or anthracite for the thermal power plant was 100 tons/yr and the calculations of carbon emission for stationary combustion were shown in Figure 4. The baseline carbon emissions of coking coal and anthracite are 268.1 and 263.7 t CO_2e/yr, respectively. When replaced with upgraded biogas, the project carbon emissions decreased to 154.1 and 145.9 t CO_2e/yr, respectively. In summary, the replacement of coking coal and anthracite by upgraded biogas for stationary combustion reduced 42.5% and 44.4% of carbon emissions, respectively. The local micro-grid system can be integrated by using upgraded biogas to replace coals and mitigate carbon dioxide emissions.

Overall, although applying a biogas desulfurization facility combined with the hollow fiber membrane technology is not an advanced idea, the combination of biogas bio-desulfurization and upgrading systems on a farm scale has never been reported.

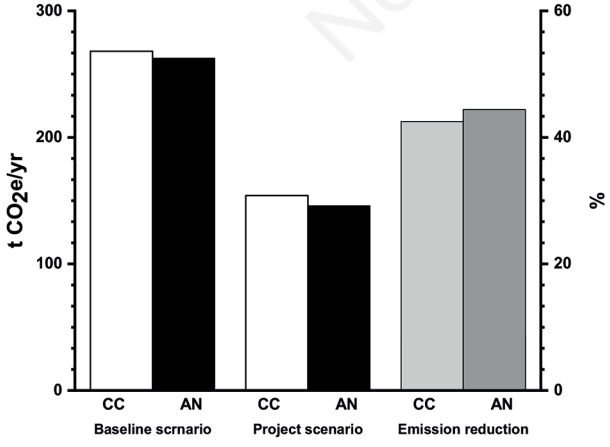

Figure 4. Comparison of the baseline scenario, project scenario, and emission reductions of carbon dioxide by replacing coking coal (CC) and anthracite (AN) with upgraded biogas for stationary combustion.

Conclusions

The study successfully demonstrated the feasibility and effectiveness of a farm-scale biogas upgrading system combined with a bio-desulfurization system. Results showed that the biogas upgrading system provided the maximum daily treatment volume of 20 L/min and achieved 100% of CO_2 removal and 94% of methane content in the treated livestock biogas. The cost-effectiveness of the upgrading system showed that a larger-scale system is more beneficial. Finally, the replacement of anthracite and coking coal by upgraded biogas for stationary combustion significantly reduced above 40% of carbon dioxide emissions. National feed-in tariffs for developing renewable energy may be varied in different countries, but they can accelerate the growth of the domestic renewable energy industry. The achievement of this study can be applied to all decentralized pig farms worldwide for biogas upgrading and replacing cocking coal or anthracite as sustainable cleaner energy by integrating local micro-grid systems.

References

Abatzoglou N, Boivin S (2009). A review of biogas purification processes. Biofuel Bioprod Bior 3:42-71.

Adnan AI, Ong MY, Nomanbhay S, *et al.* (2019). Technologies for biogas upgrading to biomethane: A review. Bioengineering 6:92.

Afrox (2022) Product reference manual. Edition 10. 10th ed.). Afrox. Available from: https://www.afrox.co.za/en/images/2022%20Product%20Reference%20Manual_tcm266-664556.pdf

Chen XY, Vinh-Thang H, Ramirez AA, *et al.* (2015). Membrane gas separation technologies for biogas upgrading. RSC Adv 5:24399-448.

Cozma P, Ghinea C, Mamaliga I, *et al.* (2013). Environmental impact assessment of high-pressure water scrubbing biogas upgrading technology. Clean-Soil Air Water 41:917-27.

COA (2021) Quantity and value of farm products (in Chinese). ASY, COA, Taiwan. Available from: https://agrstat.coa.gov.tw/sdweb/public/book/Book.aspx

COA (2022) Pig Production Report -May 2022 (in Chinese). ASY, COA, Taiwan. Available from: https://agrstat.coa.gov.tw/sdweb/public/book/Book.aspx

Elgas Ltd. (2021.) LPG properties & LPG composition –What are the properties of LPG? Available from: https://www.elgas.com.au/blog/453-the-science-a-properties-of-lpg/

Falbo F, Tasselli F, Brunetti A, *et al.* (2014). Polyimide hollow fibre membranes for CO_2 separation from wet gas mixtures. Braz J Chem Engin 31:1023-34.

FAO (2022). Production of livestock and livestock commodities, Chapter 2, Statistical Yearbook 2022. Rome, FAO. Available from: https://www.fao.org/3/cc2211en/online/cc2211en.html#chapter-2_2

Ibrahim MH, El-Naas MH, Zhang Z, Van der Bruggen B (2018). CO_2 capture using hollow fibre membranes: a review of membrane wetting. Energ Fuel 32:963-78.

IPCC (2006a). Chapter 1. Introduction, Vol. 2. Energy, 2006 IPCC Guidelines for National Greenhouse Gas Inventories.

IPCC (2006b). Chapter 2. Stationary Combustion, Vol. 2. Energy, 2006 IPCC Guidelines for National Greenhouse Gas Inventories.

IPCC (2007). Chapter 2. Changes in atmospheric constituents and in radiative forcing. In: Climate change 2007: The physical science basis. Available from: https://www.ipcc.ch/report/ar4/wg1/

McCarthy TM (1998). Nutzung von Biogas: Probleme und Loesungen fuer Spurenbestandteile. Abwasser 139:204-7.

Pugesgaard S, Olesen JE, Jørgensen U, Dalgaard T (2013.) Biogas in organic agriculture—effects on productivity, energy self-sufficiency and greenhouse gas emissions. Renew Agr Food Syst 29:28-41.

Rövekamp P, Schöpf M, Wagon F, *et al.* (2021). Renewable electricity business models in a post feed-in tariff era. Energy 213:119228.

Su JJ (2020). Mitigation of greenhouse gas emission through anaerobic digestion of livestock waste. In: L. Pawloski, Z. Litwinczuk and G. Zhou, editors. The role of agriculture in climate change mitigation. Leiden, CRC Press/Balkema; pp. 45-56.

Su JJ, Chang YC, Chen YJ, *et al.* (2013). Hydrogen sulfide removal from livestock biogas by a farm-scale bio-filter desulfurization system. Water Sci Technol 67:1288-93.

Su JJ, Chen YJ, Chang YC, Tang SC (2008). Isolation of sulfur oxidizers for desulfurizing biogas produced from anaerobic piggery wastewater treatment in Taiwan. Aust J Expl Agr 48:193-7.

Su JJ, Chen YJ, Chang YC (2014). A study of a pilot-scale biogas bio-filter system for utilization on pig farms. J Agr Sci 152:217-24.

Su JJ, Chen YJ (2015). Monitoring of sulfur dioxide emission resulting from biogas utilization on commercial pig farms in Taiwan. Environ Monitor Assess 187:1-8.

Su JJ, Chen YJ (2018). Monitoring of greenhouse gas emissions from farm-scale anaerobic piggery waste-water digesters. J Agr Sci 156:739-47.

Su JJ, Chung HC (2021). Study of livestock biogas upgrading using a pilot-scale photocatalytic desulphurizer followed by a hollow fibre carbon dioxide adsorption module. J Agr Sci 159:3-10.

Su JJ, Hong YY (2020). Removal of hydrogen sulfide using a photocatalytic livestock biogas desulfurizer. Renew Energ 149:181-8.

Su JJ, Liu BY, Chang YC (2003). Emission of greenhouse gas from livestock waste and wastewater treatment in Taiwan. Agr Ecosyst Environ 95:253-63.

Taiwan Ministry of Economic Affairs (2023). Official notice of 2023. Feed-in tariff by renewable energy (in Traditional Chinses). Bureau of Energy, Taiwan Ministry of Economic Affairs. Available from: https://www.moeaboe.gov.tw/ECW/populace/news/News.aspx?kind=1&menu_id=41&news_id=29288

Tantikhajorngosol P, Laosiripojana N, Jiraratananon R, Assabumrungrat S (2019) Physical absorption of CO_2 and H_2S from synthetic biogas at elevated pressures using hollow fibre membrane contactors: the effects of Henry's constants and gas diffusivities. Int J Heat Mass Tran 128:1136-48.

Vogler S, Braasch A, Buse G, *et al.* (2013). Biogas conditioning using hollow fibre membrane contactors. Chem-Ing-Tech 85:1254-8.

Žák M, Bendová H, Friess K, *et al.* (2018). Single-step purification of raw biogas to biomethane quality by hollow fibre membranes without any pretreatment – an innovation in biogas upgrading. Sep Purif Technol 203:36-40.