Improvements in a Membrane-separation-type Biogas Refining-compression-filling (RCF) Facility for Dairy-farming Areas in Japan

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Abstract. A method to directly use biogas as a gaseous energy source was investigated. Utilizing this method, a prototype “refining-compression-filling (RCF) facility” for biogas, which uses membrane separation, was developed. The facility forms the core of a system for supplying dairy farms and facilities in suburbs with energy by filling gas cylinders with biogas reformed to a high calorific value. The recovery rate of methane from the biogas achieved by this prototype is about 80%, and for mass production of equipment, the officials and manufacturers in municipalities introducing the facility have requested further improvement in the methane-purification efficiency of the RCF facility. The purpose of this study was to establish optimal arrangements of the membrane module unit for improving the methane-purification efficiency of the biogas-dedicated purification RCF facility. The results of this study showed that a combination of three membrane modules arranged in series (a “three-stage-module in-series type”) was found to effectively increase the methane concentration to about 99%. To further increase methane-recovery rates, a module configuration that combines the series and parallel arrangements of modules (“three-stage-module composite type”) was found to effectively increase the methane recovery rate to approximately 95%.

1. Introduction

Biogas plants installed in dairy farms in Japan are attracting attention as a kind of facility that can produce so-called “surplus biogas” as a renewable energy source. Using livestock manure as the raw material, they can produce high-quality fertilizer (digested dairy slurry) while suppressing ammonia volatilization during processing, reduce greenhouse-gas (GHG) environmental load, and produce the biogas required to maintain the temperature of anaerobic fermenters [1]. It is possible to convert the surplus biogas into (i) heat energy in the form of hot water for washing cattle sheds in the dairy farm where the biogas plant is installed or (ii) electrical energy in the form of electricity generated by a gas generator [2][3]. However, the use of converted energy in Japan dairy farms faces problems with heat utilization and selling electricity for two reasons: first, the main facilities are scattered around the country and dairy farms are far apart and, second, the power grid is vulnerable. Specifically, dairy farms and houses (homes and buildings) with high demand for hot water are scattered about dairy-farm areas, and as is the case in northern Europe, installing pipelines for hot water and transferring heat within those areas necessitates a substantial investment, making it desirable to limit the use of hot
water at farms at which biogas plants are installed. Compared to hot water, electricity does not require investment in initial infrastructure because it can utilize the existing electric grid of the power utilities. However, in 2012, a law concerning “feed-in tariffs” (FITs)—which obliges electric-power companies to purchase electricity generated by using renewable energy sources—has been implemented. Even so, due to the low power-transmission capacity in dairy farming areas, it is impossible for some electric-power companies to sell electricity due to restrictions on their electricity-purchase quota. For these reasons, to promote effective use of biogas in dairy farms, it has become desirable to establish a method for directly transporting biogas, which is subject to low losses during the energy reforming, to be used in areas outside of the location of the operating system generating the gas [4][5].

A method of directly using biogas as a gaseous energy source was investigated. Utilizing this method, a prototype refining-compression-filling (RCF) facility for biogas, which uses a gas-separation membrane, was developed. The facility forms the core in a system for supplying dairy farms and facilities in the suburbs by filling gas cylinders with biogas reformed to high calorific value [6]. The prototype was operated on a test basis in “Town A” in Hokkaido, Japan. With a single-stage membrane module, it achieved a basic purification efficiency of about 80%. To enable mass production of the prototype, however, it was pointed out that the methane-purification efficiency of the refining compressor should be further improved.

Accordingly, the purpose of this study was to identify an optimal arrangement of the membrane module for improving the methane-purification efficiency of the biogas-dedicated purification part of the RCF.

2. Methods

2.1. Improvements in the equipment
For the operational test of the prototype, it was shown that (i) the constant-temperature control of the membrane module was unstable in the severely cold season and (ii) the dehumidification of the biogas was unstable in the summer as well as in the severely cold season. To solve these two problems, the following improvements were made to the equipment, and the arrangement of membrane modules was varied and tested.

1) To improve the gas-purification efficiency, a gas cooler (chiller unit) using water cooling was introduced. The chiller unit was used to rapidly cool the gas after pressurization and generate condensed water, which was then used for stable dehumidification.
2) Based on a heat exchange between the compressed and cooled gases and an electric heater, an arrangement to raise the temperature of the cooled gas to the optimum temperature for the methane-separation membrane (about 50 °C) was constructed.
3) A unit combining a dehumidifying function and methane separation was fabricated, the unit was integrated with the control system, the gas piping was simplified, and as a result, the entire facility could be downsized.

2.2. Investigation of the arrangement of membrane modules
With the improved test equipment, a test optimizing the arrangement of the membrane module was conducted. The test was conducted on surplus biogas generated from a biogas plant (with a raw-gas supply by 250 head of livestock) in Town A. The arrangement of the membrane module and the test conditions—alongside the biogas-purification flow used in each test—are shown in Figure 1. The particulars of the test biogas-dedicated purification part are outlined in Figure 2.

In the tests, 4 types of membrane modules, one stage (test 1), two stages (test 2) (“series arrangement”), and three stages (“series arrangement (test 3)”; as well as a “composite arrangement (test 4),” a series and parallel composite arrangement), were tested. Preliminary tests using one or two
stages (tests 1& 2) of the membrane module were conducted to determine the methane concentration and recovery rates.

Figure 1. Biogas-purification flow used in Test 1-Test 4.
In the test of the three-stage membrane module, a test system that can be arranged in series (“three-stage series type”) or in combination (“three-stage composite type”) was configured by switching piping and valves. The gas-separation membrane was a hollow fiber membrane type (Air Products and Chemicals, Inc., Allentown, USA). For the biogas-purification test, at the optimum purification conditions for the membrane-module unit of the developed purification equipment (temperature: 50 °C (Limit of heat resistance of membrane module); pressure: 0.6 MPa; flow rate: 5 m$^3$/h), the methane concentration, pressure, and flow rate were measured, and the methane recovery rate was calculated. The methods for measuring each parameter are as follows.

1) Methane concentration
For the investigation of gas components, a gas analyzer (GA2000 manufactured by Geotech, UK) was connected to the sample valves, and the methane concentration was determined. The methane concentration of the refined gas was the value measured at point ⑦ (Figure 2, #3 Refined biogas (rich-gas)) for the three-stage series type and at point ⑤ (Figure 2, #4 rich-gas) for the three-stage combined type.

2) Pressure and flow rate
The pressure was measured by manually reading off a pressure gauge installed near each sample valve. For the flow rate, some of the measurement points are not fitted with a flow meter; and a flow meter was therefore attached to these sample valves by using a pipe for the measurements. The flow meter used here is compatible with gas at a pressure of 0.6 MPa and composition of CH$_4$: CO$_2$ = 60%:40%, the values were corrected after the measurement.

3) Methane-recovery rate
The methane-recovery rate was calculated by using formula (1) below, and based on the methane concentration and flow rate of the raw untreated gas and methane concentration and the flow rate of the purified gas:

$$\text{Recovery rate} = \frac{(\text{CH}_4 \%) \text{ of purified gas} \times \text{flow rate of purified gas}}{\text{CH}_4 \% \text{ of raw material gas} \times \text{flow rate of raw material gas}} \times 100 \ldots (1)$$
3. Results and Discussion

3.1. Purification test of biogas-purification equipment with the built-in hybrid membrane module

(1) Preliminary test; Test with single-stage and two stage membrane module (Tests 1, 2)

1) Test with the single-stage membrane module (Test 1)
   The single-stage purification test (using one membrane module) was executed by a single membrane module with different biogas-purification capacities. The results of the test showed that the methane concentration was increased by decreasing the purified-gas production capacity. However, if the production capacity is reduced to 2 m³/h, and the methane concentration rises to 98.0%, the amount of permeated methane in the off-gas (the separated gas with low methane concentration) increases, making the methane loss increase and the methane-recovery rate drops to 25%. This result shows that the membrane module with a production capacity of 5 m³/h achieves the highest methane-recovery rate.

2) Test with the two-stage membrane module (Test 2)
   When two membrane modules (with production capacity of 5 m³/h) were placed in series (two-stage series) and tested, methane concentration in the purified gas was found to be 97%. However, it was also found that the methane-recovery rate was about 50% because the off-gas from each stage contained a large amount of methane. Given this result, recovery of the methane was attempted by restoring the off-gas from the first-stage membrane module to the raw-material gas; however, because the methane concentration in the off-gas was very low, the result was to lower the methane concentration in the purified gas. Therefore, when only the off-gas from the two-stage membrane module was restored to the raw gas, the methane-recovery rate improved to 88%; however, the concentration of methane in the purified gas at that time dropped to 95%.

   From the above test results, it was concluded that it is effective to use a three-stage membrane module to further increase the methane concentration and methane-recovery rate. As a means of arranging the membrane module, a three-stage series (three membrane modules arranged in series) and a three-stage composite type (three membrane modules arranged in series and in parallel) were devised, and the following tests were carried out.

(2) Test with the three-stage series type apparatus (Test 3)
   The measured results of methane and carbon-dioxide concentrations, gas-flow rate, and other parameters with the three-stage series type arrangement are listed in Table 1. In the raw-gas purification test, methane concentration and carbon-dioxide concentration of the raw-gas were 59.7% and 40.0%, respectively, and the methane and carbon-dioxide concentrations of the purified M-3 rich-gas were 98.9% and 1.0%, respectively. It was considered that the flow rate of the M-1 off-gas, which is the off-gas released to the atmosphere, was almost at the setting value, and the nitrogen remaining in the equipment was eliminated. As this result, the methane concentration of the M-3 rich gas increased to 98.9%; however, the methane-recovery rate decreased to 81.2% owing to the decrease in the flow rate. The flow rate of the M-3 rich gas, which is the final purified gas, was 4.9 m³/h at a raw-gas flow rate of 10.0 m³/h (Test 3 in Table 1).
Table 1. Measured values for the three-stage series type (test 3).

| Measured value*1 | Raw-gas | CP outlet*5 | M-1 rich-gas | M-1 off-gas | M-2 rich-gas | M-2 off-gas | M-3 rich-gas | M-3 off-gas |
|------------------|---------|------------|--------------|-------------|--------------|-------------|--------------|-------------|
| CH₄ concentration (%) | 59.7 | 59.7 | 81.4 | 17.5 | 96.0 | 42.3 | 98.9 | 86.3 |
| s.d.*2 | 0.4 | 0.4 | 0.8 | 0.7 | 0.6 | 0.6 | 0.4 | 0.6 |
| CO₂ concentration (%) | 40.0 | 40.0 | 17.8 | 81.7 | 3.3 | 57.2 | 1.0 | 12.7 |
| s.d.*2 | 0.3 | 0.3 | 0.5 | 0.5 | 0.2 | 0.5 | 0.2 | 0.5 |
| Pressure (MPa)*3 | 0.0 | 0.6 | 0.6 | 0.0 | 0.6 | 0.0 | 0.6 | 0.0 |
| Flow rate (m³/h) | 10.0 | 16.4 | 10.7 | 5.1 | 7.6 | 3.4 | 4.9 | 2.6 |
| Recovery rate (%)*4 | — | — | — | — | — | — | 81.2 | — |
| Percentage of raw-gas (%)*4 | 100.0 | — | — | 51.0 | — | — | 49.0 | — |

*1 Operating conditions: Flow rate: 10 m³/h; pressure: 0.6 MPa; temperature: 50.0°C
*2 Standard deviation of concentration
*3 Pressure: Gauge pressure value
*4 Ratio of purified-gas amount and off-gas amount to raw-gas amount
*5 CP outlet: compressor outlet,

(3) Test with three-stage composite type apparatus (Test 4)
The measured results of methane and carbon-dioxide concentrations, gas-flow rate, and other parameters with the three-stage composite type arrangement are listed in Table 3. In the purification test with the three-stage composite type, the methane and carbon-dioxide concentrations of the raw gas were 59.7% and 40.0%, respectively, and the methane and carbon-dioxide concentrations of the purified gas were 95.6% and 4.3%, respectively. Since the flow rate of the raw-gas reached the set value (5 m³/h), the flow rate of the M-3 off-gas, which is the off-gas released to the atmosphere, was improved. As a result, the methane concentration of the purified M-2 rich-gas increased to 95.6%, and the methane-recovery rate increased to 94.5% owing to the increased gas-flow rate. The flow rate of the raw-gas was 10.0 m³/h, whereas the flow rate of the final purified methane gas (M-2 rich-gas) was 5.9 m³/h (Test 4 in Table 1).

Table 2. Measured values of gas parameters in the three-stage composite type arrangement (test 4).

| Measured values*1 | Raw-gas | CP Outlet*5 | M-1 rich-gas | M-1 off-gas | M-2 rich-gas | M-2 off-gas | M-3 rich-gas | M-3 off-gas |
|------------------|---------|------------|--------------|-------------|--------------|-------------|--------------|-------------|
| CH₄ concentration (%) | 59.7 | 59.7 | 75.6 | 2.8 | 95.6 | 52.7 | 55.1 | 2.8 |
| s.d.*2 | 0.4 | 0.4 | 1.1 | 0.3 | 0.7 | 0.6 | 0.4 | 0.3 |
| CO₂ concentration (%) | 40.0 | 40.0 | 25.6 | 96.7 | 4.3 | 45.9 | 48.3 | 96.1 |
| s.d.*2 | 0.3 | 0.3 | 0.6 | 0.5 | 0.1 | 0.5 | 0.2 | 0.4 |
| Pressure (MPa)*3 | 0.0 | 0.6 | 0.6 | 0.2 | 0.6 | 0.0 | 0.15 | 0.0 |
| Flow rate (m³/h) | 10.0 | 14.5 | 10.0 | 4.6 | 5.9 | 3.7 | 2.5 | 4.1 |
| Recovery rate (%)*4 | — | — | — | — | — | 94.5 | — | — |
| Percentage of raw-material gas (%)*4 | 100 | — | — | — | 59 | — | — | 41 |

*1 Operating conditions: Flow rate: 10 m³/h; pressure: 0.6 MPa; temperature 50.0 °C
*2 Standard deviation of concentration
*3 Pressure: Gauge pressure value
*4 Ratio of purified-gas amount and off-gas amount to raw-gas amount
*5 CP outlet: compressor outlet,

The results of the biogas purification tests (Tests 3 and 4) are detailed in Table 4. The methane concentration of the purified gas after the purification process is 95% or better in all of the methods (membrane-module arrangements). The methane-recovery rate was 94.5% with the three-stage
composite type, compared to 79.0% [6] for the single-stage membrane module; an increase of 15.5 points (about 20%) over that of the former method.

In the developed device, we believe that the introduction of a chiller unit to cool the compressed gas enabled sufficient dehumidification and improved the methane separation performance of the membrane module. With the methane concentration, about 1 to 2% of air is mixed in because oxygen is required to promote the activity of sulfur sulfide bacteria in biological desulfurization, but oxygen and nitrogen are sufficiently separated by the membrane module, so only methane remains. It is considered that the methane concentration has improved.

These test results showed that the three-stage series type is suitable for obtaining a purified gas with a high methane concentration, while the three-stage composite type is suitable for increasing methane-recovery rates. It is concluded that when the purified gas is used locally, outside the operation system where it has been concentrated, increasing the methane concentration may improve adaptability to commercial gas equipment and reduce delivery costs; when the purified gas is used at the location of the operation system, the three-stage composite type (with high methane-recovery efficiency) is suitable, because utilization efficiency is superior.

Table 3. Details of results (biogas-purification test).

| Item: Number of stages/membrane arrangement | CH4 concentration (%) | CO2 concentration (%) | Methane-recovery rate*3 (%) |
|---------------------------------------------|-----------------------|-----------------------|-----------------------------|
| Raw-gas*1                                   | 55–60                 | 40–45                 |                             |
| Purified gas*2,*5                           | Protocol facility*4    | 94.5                  | 5.5                         | 79.0                         |
|                                             | Three-stage series type (test3) | 98.9 | 1.0 | 81.2 |
|                                             | Three-stage composite type (test 4) | 95.6 | 4.3 | 94.5 |

*1 Raw-gas: Biogas to be purified
*2 Purified gas: Gas with methane concentration increased by the purification process
*3 Methane-recovery rate: Ratio of methane gas contained in purified gas to methane gas contained in raw gas
*4 Prototype facility: Existing refining-compression-filling facility [6]
*5 Refining conditions: temperature: 50°C; pressure: 0.6 MPa; raw-material flow rate: 10 m³/h

4. Conclusions
This study was aimed at establishing the optimal arrangement of the membrane module for improving the methane-purification efficiency of the biogas-dedicated purification part of an RCF.

From the test results, producing purified methane by using a membrane module, it was found to be effective to use a three-stage membrane module to increase methane concentration and the methane-recovery rate. These test results showed that a three-stage series type is suitable for obtaining a purified gas with high methane concentration, while the three-stage composite type is suitable for increasing the methane-recovery rate. It is concluded that when the purified gas is used locally, away from the operation system where it is generated, the three-stage composite type with high methane-recovery efficiency is suitable.

However, since the methane recovery rate may be affected by the off-gas pressure of the membrane modules at each stage, it is possible to consider conditions for further increases in the methane recovery rate, such as changing the pressure conditions between the membrane modules. It is considered that it will be possible to approach the expected value (purification efficiency CH4 concentration; 90% or more). The inflow temperature of the raw-gas into the membrane module depends on the heat resistance limit of the membrane module. From these findings, it is considered that there is room for consideration of the number of membrane stages, the arrangement method, and the materials composing the hollow fiber membrane and the whole of the module.

In this way, refined gas obtained from the surplus gas of individual biogas plants introduced for the purpose of dealing with livestock manure and reducing the environmental load can be regarded as
one of the distributed energy sources in agricultural and rural areas, and it is possible to develop local demand destinations. For the future, it is necessary to continue technological development such as adaptation technology to existing supply and consumption facilities, efficient transportation, and the delivery technology.

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