



Methane enrichment of biogas by carbon dioxide fixation with calcium hydroxide and activated carbon



Muhammad Rashed Al Mamun^a, Mohammad Razaul Karim^{b,*}, Mohammed M. Rahman^c,
Abdullah M. Asiri^c, Shuichi Torii^a

^a Department of advanced Mechanical System Engineering, Graduate school of Science and Technology, Kumamoto University, 2-39-1 Kurokami, Kumamoto, 860-8555, Japan

^b Department of Chemistry, School of Physical Sciences, Shahjalal University of Science and Technology, Sylhet 3114, Bangladesh

^c Center of Excellence for Advanced Materials Research (CEAMR) & Chemistry Department, King Abdulaziz University, P.O. Box 80203, Jeddah 21589, Saudi Arabia

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ABSTRACT

Although, Biogas generated from anaerobic digestion process is a clean, carbon neutral and environmental friendly energy alternative, raw biogas needs to be purified necessarily before using. The removal of CO₂ from raw biogas ensures its better applications in engines, heating purposes and electricity generation. Reduction of CO₂ content also enhances the calorific value. Biogas generated from vegetable, fruit and cafeteria wastes for duration of 30 days with the average concentration for CH₄ and CO₂ as 62 and 34%, respectively have been observed. The CO₂ content was possible to reduce significantly by treating the raw biogas with purifying agents as: solid CaO, CaO solution and activated carbon. Purifying agent's mass and concentration and treatment time dependent biogas upgradation profile reveals that these novel purification processes are effective and affordable.

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1. Introduction

In recent years the issues on climate change, increasing dependency on fossil fuels and increasing cost for electricity generation have awakened a global concern for greater shares of renewable energy in future power supplies [1]. As the natural processes for formation of fossil fuels are extremely slow (hundreds to thousands of years duration), there exists no possibility for natural regeneration of such fuels [2]. In addition, the green house gas emissions related to energy production from fossil fuels is a cognizant issue. Therefore, attempts for finding green, sustainable and renewable alternatives to fossil fuels have attracted worldwide interests. In this respect, various means for efficient conversion of biogas into energy have become popular. The main sectors associated with biogas oriented applications includes: electricity generation, heating purposes and transportation [3]. Biogas production from biomass is one of the cheapest and most fascicle technical pathway for alternative fuel generation [4]. The ease for collecting biomass for biogas generation and the direct utilization of biogas to operate agriculture machineries indicate its immersing possibility in farm sectors or rural areas [5]. As an additional advantage, biogas generation facilitate the solid waste management, which

is a major necessity in urban life [6]. Biogas is a flammable mixture of different gases, which are produced by decomposition of biodegradable organic matters in cattle dung, vegetable wastes, sheep and poultry droppings, municipal solid wastes, industrial wastewater and so on by anaerobic action of microorganisms.

The composition of biogas depends on the type of raw materials and on the temperature, pressure and pH inside the reaction tank [7]. An approximate composition of biogas includes: CH₄ (50 to 70%), CO₂ (30 to 40%), H₂ (5 to 10%), N₂ (1 to 2%), H₂O (0.3%) and trace amount of H₂S [8]. The high calorific value of CH₄ (37.78 MJ/m³) makes biogas a good renewable energy source [9]. The biogas calorific efficiency is proportional to the CH₄ concentration, while for using in internal combustion engines, the minimum concentration of CH₄ in biogas should be 90% [10]. The large volume of CO₂ is undesirable, as it reduces the calorific value of biogas, increases the compression and transportation costs and limits the economic feasibility. Furthermore, the presence of carbon dioxide in biogas can cause problem by freezing at the flow control valves and metering points. Therefore, purification of biogas by minimizing CO₂ and maximizing CH₄ is necessary for its efficient application for heating purposes, electricity generation and as fuels in vehicle or engines [11].

Todate, some common methods for removing CO₂ from biogas includes: physical and chemical adsorption, pressure swing adsorption (PSA), membrane based separation, biological or chemical fixation of CO₂, cryogenic separation and so on [12–27]. In some

* Corresponding author.

E-mail address: krazaul@yahoo.com, mrzaulk-che@sust.edu (M.R. Karim).

chemical treatment process the solvents mono-ethanolamine (MEA), di-ethanolamine and tri-ethanolamine or aqueous solution of alkaline salts (*i.e.* sodium, calcium hydroxide and potassium) are being used [12,13]. Some water scrubbing system employing a gas inlet pressure and flow rate of 1.0 MPa and 1.5 m³/h, respectively with water flow rate as 1.8 m³/h could reduce the CO₂ content by 99% [28]. Biswas et al. [29] could decrease the CO₂ content from 40 to 0.7% by bubbling the raw biogas through 10% solution of MEA. An important advantage of using MEA is that it can be reutilized after boiling five minutes. The use of alkaline solution of KOH, or NaOH for the removal of CO₂ is well established phenomenon, where the agitation enhances absorption of CO₂ by aiding homogenous diffusion of CO₂ molecule in the solutions. However, CO₂ absorption is found to be the most rapid in 2.5 to 3.0 mol/dm³ NaOH [30]. The water scrubbing method could purify CH₄ up to 100% through counter current flow of the gas and water solution [4]. CO₂ content was possible to reduce from 30 to 2% by employing a flow rate of gas and water as 1.8 and 0.465 m³/h, respectively [31]. In another study, at 48 kPa pressure with a water flow rate of 2 m³/h could result in the 87.6% removal of CO₂ [32].

CO₂ have almost 20 times higher permeability than CH₄ through solid phase membrane based on acetate–cellulose polymer. Using such membrane the biogas mixtures can be separated by applying 2500 to 4000 kPa pressure [33]. Monsanto and acetate cellulose membranes result in high removal efficiency for CO₂ [34]. Adsorption based purification using naturally occurring solid Zeolite Neopoliton Yellow Tuff (NYT) is also common [35]. Polyvinyl amine (PVA) on different carriers and cross linkers also can be used for removal of unwanted gas. Typical PVA on polysulfone cross linked with ammonium fluoride exhibits the highest selectivity for CO₂ compared with CH₄ (>1:1000) [24]. Though the membrane module enhances the separation performance and high purity CH₄ can be obtained, in some cases the separation efficiency is rendered due to the poisoning effect and sometimes the losses of CH₄ is significant. The corrosion of PSA by H₂S is a common example [16,20,33]. In cryogenic separation, using fractional condensation/distillation of gas mixture at low temperature CH₄ concentration up to 97% can be availed [23]. One advantage of this process is the possibility of recovering pure components in the form of liquid. However, complicated flow streams and low thermal efficiency is unfeasible and requires high costing [36].

All the as mentioned biogas purification and CH₄ upgradation processes are well studied and associated with their own advantage and disadvantages. In spite, it is necessary to find out some non expensive and viable biogas purification methods, which could be applied in small farm sectors in rural area. Considering these issues, the aim of this project is to use commercially available solid CaO, CaO solution and activated carbon for removing carbon dioxide from biogas generated by anaerobic co-digestion process. We report the continuous flow methods for nonstop purification of raw biogas. The cheap and inexpensive nature of both the chemical agents suggests the possible utilization of raw biogas through purification in small scale plants.

2. Materials and methods

2.1. Collection of fermentation materials

The vegetable and fruit wastes were collected from kokai vegetable market, Kumamoto, Japan. Cafeteria wastes were collected from the cafeteria of Graduate School of Science and Technology at Kumamoto University. The materials were stored at 4 °C before charging into the gas generation tank.

2.2. Production of biogas

A laboratory scale plant was assembled for production and purification of biogas. It contains a digester, collection chamber, gas

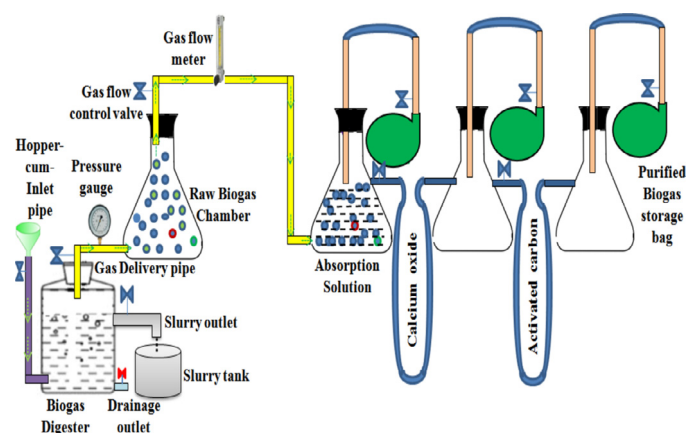


Fig. 1. Experimental setup for generation of biogas and removal of CO₂.

purification unit and gas storage system. The experimental setup for the gas production and purification is presented in Fig. 1. The 0.2 m³ polypropylene digester tank (used as a reactor) with an air tight rubber gasket was interconnected to the purification unit. Cafeteria, vegetable and fruit wastes were cut into small pieces and were mixed in equal proportion. A homogenous mixture was obtained, which was diluted with tap water at 1:1 ratio. In order to optimize gas production, feed stock was maintained to contain 8% of total solid. The prepared raw materials were used as input charge for the production of biogas in the digester tank. About 80% of the digester volume was filled with the diluted organic waste. For better digestion, the batch pH was maintained between 6.8 and 7.6 by adding lime (CaO). Mesophilic batch digestion was conducted at 33 ± 2 °C temperature under 30 days hydraulic retention time. Inside the digester, biogas was generated from anaerobic decomposition of the input sample by the action of various methanogenic microorganisms and bacteria. The amount of biogas generated was monitored for 30 days.

2.3. Design of purification unit

In purification, we intended to enrich the CH₄ content in the biogas by removing undesired CO₂. The spontaneous pressure and constant flow rate (0.0025 m³/min) of biogas produced inside the digester was utilized to flow the gas through air tight U tubes filled by variable amount of CaO or activated carbon. We also repeated the experimentation for removal of CO₂ by passing the raw biogas through CaO solution in water of varied concentration. The purification unit consists of three 0.002 m³ glass flask connected through 6 mm and 13 mm hose pipe with flow control valves. The 1st flask was filled by CaO solution, whereas the 2nd and 3rd one was filled by biogas which was flowed inside before starting the purification process. There are two U shaped hose pipes having 13 mm diameter and 1.5 m length to convey the gas. The hose pipes were filled with CaO and activated carbon powder of varied quantity (from 1 to 10 g each) in different experiments. Under batch type operational conditions, the biogas was introduced at the bottom of the absorber flask as small bubbles. The gas passed through the flasks and U tubes. Finally, the purified gas was stored in a gas bag for sampling in gas chromatography. A series of sample were collected from raw biogas and that one purified passing through varied quantity of CaO and activated carbon. For time dependent concentration measurement, 1 L of raw biogas was flowed through 5 g each of the adsorbed phase inside a U tube. Sets of experiments were repeated by pressing one the liter gas through the tube with various time durations ranging from 2 to 24 h. In this step the gas flow rate was controlled by external pressure.

To obtain and detect the best results we repeated each experimental step by three times. The best data obtained from the gas

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