

# A new approach for upgrading biogas produced at a local wastewater treatment plant

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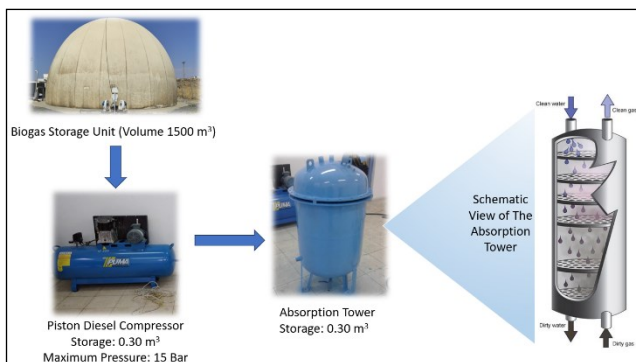
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## Graphical abstract



## Abstract

Biogas is globally recognized as a crucial source of renewable energy. After undergoing proper treatment, the purified form of biogas, known as "biomethane", has the potential to be injected into existing natural gas infrastructure or compressed for use as a vehicle fuel. The principal pollutants which cause a reduction in the energy density of biogas and impede its utilization, specifically in engines, are carbon dioxide, hydrogen sulfide, and siloxanes. Furthermore, the presence of carbon dioxide in biogas has a significant impact on global warming. The water scrubbing method, which is the most used method in biogas upgrading processes, was utilized due to its low operating cost. With the decreasing availability of clean water resources around the world and the challenges of utilizing them, the reuse of treated wastewater has become increasingly important. Therefore, in this study, treated wastewater was used as scrubbing water for biogas instead of clean water. In addition, the biogas used in the study is the gas produced in the wastewater treatment plant of a local municipality, thus contributing to the uniqueness of the study. Prior to upgrading, the biogas content produced in the plant had CH<sub>4</sub> and CO<sub>2</sub> values of 62% and 37%, respectively. However, after the implementation of upgraded technology, these values increased to 96% (CH<sub>4</sub>) and 3% (CO<sub>2</sub>). These results clearly demonstrate the success of the new method.

**Keywords:** Biogas upgrading, CO<sub>2</sub> removal, water scrubbing

## 1. Introduction

With the increasing demand for energy and the threat of global warming, there is a critical need to search for alternative energy sources. Currently, natural gas, oil, and coal account for about 80% of the world's energy consumption due to their high energy value and availability (Qyyum *et al.* 2020). Most renewable energy sources generate power at varying levels during different times of the day and season. However, energy from biomass can be consistently stored and produced. Biomass resources are obtained from organic sources such as agricultural waste, food waste, sludge from municipal treatment plants, and animal manure (Pattharaprachayakul *et al.* 2021). Overall, biomass can be converted into biogas through the process of anaerobic fermentation. "Anaerobic digestion is a multi-stage process that involves hydrolysis, acidogenesis, acetogenesis/dehydrogenation, and methanation (Mergenthal *et al.* 2022). While biogas is mainly used for generating electricity and heat through cogeneration, some of the drawbacks of this method (such as low electrical efficiency, high maintenance, and inefficient heat utilization) have prompted the advancement of alternative methods for biogas recovery (Wantz *et al.* 2022). Methane is the main component of naturally occurring biogas and has a significant impact on global warming. It has become one of the most widely used fuels for power generation, heating, and transportation over the last century. As the majority of methane used by societies today comes from natural gas, there is a growing interest in collecting methane from decaying biomass. However, this does not include capturing or collecting methane from all natural sources. Instead, it involves building dedicated biogas plants, where the biogas process can be fully contained, controlled, and optimized, allowing for a commercially viable industry to be established. This harnesses nature's potential to produce renewable and environmentally sustainable gaseous biofuels (Pattharaprachayakul *et al.* 2021).

Raw biogas is composed of 40-75% CH<sub>4</sub>, 25-60% CO<sub>2</sub>, and other gases such as H<sub>2</sub>S, N<sub>2</sub>, H<sub>2</sub> and CO (Rotunno *et al.* 2017). While biogas production is widespread and easy, its use without purification is very limited due to the corrosive nature of H<sub>2</sub>S and the high level of CO<sub>2</sub> (Ryckebosch *et al.*, 2011). However, if the CH<sub>4</sub> content of biogas is increased to at least 90%, it can be used as fuel for vehicles that run on natural gas, compressed natural gas (CNG), and liquefied natural gas (LNG). Additionally, since biogas is produced locally, there is no need for long-distance transportation, which is necessary for fossil fuels (Scarlat *et al.* 2018).

It is crucial to determine the economic and sustainable methods for separating CO<sub>2</sub> and H<sub>2</sub>S gases in biogas. A variety of technologies for biogas upgrading have been explored, including pressure swing adsorption, high pressure water scrubbing, membrane separation, cryogenic upgrading, and absorption processes (which can involve physical absorption or chemical reactions) (Ardolino *et al.* 2021).

Water Scrubbing (WS) is the most mature and widely used method, accounting for 35.5% of all existing biogas upgrading plants as of 2016 (Reck *et al.* 2018). WS has numerous advantages, including environmental friendliness, a low methane loss rate, and simple operation. However, the low solubility of CO<sub>2</sub> in water means that large amounts of water need to be circulated, requiring a large apparatus. Additionally, there are some disadvantages, such as excessive water usage, foam issues, the formation of bacterial flocs, and an increase in H<sub>2</sub>S content in the wash water, which can damage the pumps. Therefore, it is necessary to improve the efficiency of the process by proposing and/or developing new strategies (Scarlat *et al.* 2018).

The water Scrubbing method was utilized in the completion of the study. This method involves using an absorption tower to wash the biogas by spraying water from above. The water scrubbing technique separates CO<sub>2</sub> and CH<sub>4</sub> by utilizing water as an absorbent. CO<sub>2</sub> is extracted from the biogas through washing with high pressure water. The biogas is introduced from the bottom of a vertical column at a pressure of 8-10 bar, while the water is introduced from the top to create a gas-liquid mixture.

For the first time in this study, treated wastewater was used instead of clean water for the washing process in the biogas purification process. The study observed the efficiency of this method in biogas purification, as well as its effects on parameters such as pH, chemical oxygen demand (COD) and alkalinity conductivity in the treated wastewater. The washing process was carried out over a long period of time, and graphs and tables were created to show the relationship between the absorption efficiency of the wash water and the duration of usage. The resulting purified biogas was measured after the completion of the study. Additionally, the biogas used is produced in a real wastewater treatment plant, which adds original value and contributes to the existing literature.

## 2. Materials and methods

### 2.1. Siverek wastewater treatment plant

The biogas used in this study was obtained from the municipal wastewater treatment plant located in the Siverek district of Sanliurfa province. The wastewater coming to Siverek Wastewater Treatment Plant is of domestic quality and its source is households in Siverek district and water from rain drainage lines. The sludge obtained from the wastewater coming to the plant is kept in the digester and heated and mixed at the same time. After about 25 days, the sludge in the digester ferments and biogas is produced. The biogas produced is converted into electricity and heat energy by burning in cogeneration units to meet the plant's own electricity and to be used for heating the process and administrative building. The sludge, which is completely fermented in the digester, is used as agricultural fertilizer after it is thickened in machines called decanters until the dry matter ratio reaches approximately 30%. All necessary legal permits were obtained for its use.

### 2.2. Biogas upgrading

The biogas produced at the facility is stored in a storage tank with a capacity of 1500 m<sup>3</sup>. The biogas content produced in the plant is regularly measured using a gas meter (Dräger X-am 7000). The concentrations of gases present in the biogas are listed in Table 1.

**Table 1.** Biogas content produced at Siverek wastewater treatment plant

| Constituents     | Unit of Measure     | Mean | Standard Deviation |
|------------------|---------------------|------|--------------------|
| CH <sub>4</sub>  | %                   | 62.0 | ± 2.0              |
| CO <sub>2</sub>  | %                   | 37.0 | ± 1.8              |
| NH <sub>3</sub>  | mg Nm <sup>-3</sup> | 7.1  | ± 2.3              |
| H <sub>2</sub> S | mg Nm <sup>-3</sup> | 3.2  | ± 0.8              |
| Mercaptans       | mg Nm <sup>-3</sup> | 0.8  | ± 0.1              |
| Alkyl-sulfides   | mg Nm <sup>-3</sup> | 4.2  | ± 0.9              |
| Temperature      | °C                  | 55   | ± 2.9              |

The biogas used in the study was obtained from this storage tank with the help of a compressor (Piston Diesel Compressor, Storage: 0.30 m<sup>3</sup>, Maximum Pressure: 15 Bar). The biogas was then delivered from this compressor to the absorption tower (Storage: 0.30 m<sup>3</sup>) for biogas upgrading at a determined pressure. An aqua-flex diffuser (Aqua-flex

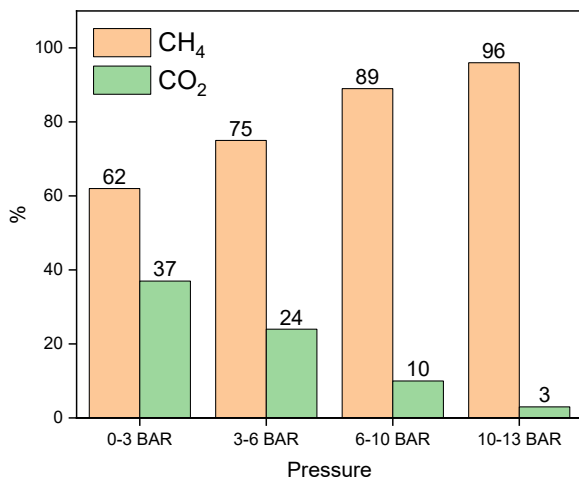
Add230-9") was used to evenly distribute the biogas on the bottom surface of the tower. A schematic representation of the absorption tower is shown in the Graphic Abstract. From the part labeled as 'clean water' in the Graphical Abstract, the 'wastewater' treated in the plant is given as a method applied for the first time, which is the unique value

of this study. In addition, pH, Alkalinity and COD analyses were performed on the water to observe the changes in the character of the treated wastewater after its use in purification. Standard methods method number; Determination of Chemical Oxygen Demand (COD) Open Reflux - Titrimetric Method TS 2789, Alkalinity Determination Titrimetric Method SM 2320 B, pH Determination Electrometric Method TS EN ISO 10523

### 3. Results and discussion

#### 3.1. CO<sub>2</sub> removal from biogas

The temperature and biogas flow rate used for the experiment were kept constant at 10 L min<sup>-1</sup>. The study was carried out by observing changes in the volume of water in the absorption tower and the outlet gas over time while keeping the volume of water in the tower constant. In the first step, emphasis was placed solely on the purification efficiency of the pressure on the biogas, while ignoring changes in the water level in the tank. In the second step, changes in the water level in the tank over time were examined under ideal pressure conditions. Since the biogas is first treated for H<sub>2</sub>S in the desulfurization unit during the upgrading process, only CH<sub>4</sub> and CO<sub>2</sub> data were used for the study.



**Figure 1.** Percentage change of CH<sub>4</sub> and CO<sub>2</sub> gases after biogas upgrading (20 °C, 10 L min<sup>-1</sup>)

Figure 1 shows the absorption of CH<sub>4</sub> and CO<sub>2</sub> at a constant temperature of 20 °C, a constant flow rate of 10 L min<sup>-1</sup> and a pressure range of 0 to 13 bar.

Upon examining the CH<sub>4</sub> concentration, it is observed that the absorption efficiency is lowest at approximately 60% in the 0-3 bar pressure range. The efficiency of CH<sub>4</sub> absorption then increases linearly until the pressure reaches approximately 9 bar. Beyond the 9 bar pressure point, it was determined that the CH<sub>4</sub> yield continued to increase linearly, but at a slower rate.

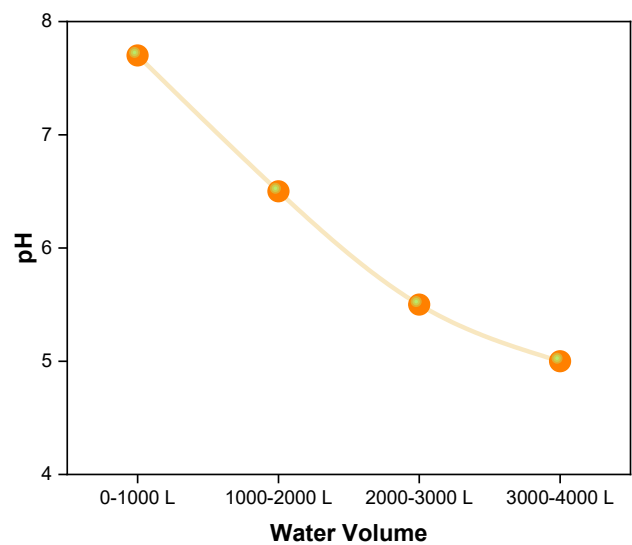
Looking at the CO<sub>2</sub> concentration, it can be seen that the absorption efficiency is highest at around 40% within the 0-3 bar pressure range. As the pressure increases up to approximately 8 bar, the absorption efficiency of CO<sub>2</sub> decreases to around 3%. Afterward, with further increases in pressure, it is observed that the CO<sub>2</sub> absorption efficiency decreases at a slower rate after 3%.

The most cost-effective pressure value was found to be 10 bar, as shown in Figure 1. At this pressure, the CH<sub>4</sub> concentration in the biogas reaches a level comparable to that of natural gas, allowing the compressed gas to be used as a vehicle fuel (Bio-CNG).

#### 3.2. Characteristics of the wastewater used for water scrubbing method.

##### 3.2.1. pH

Please refer to Figure 2, which shows the pH values at a constant temperature of 20°C and within a volume range of 0 to 4000 L. From the graph, it is apparent that the maximum value is around 7.7 at 500 L. Subsequently, the pH value gradually decreased until reaching approximately 2500 L. Beyond this point, it was noted that the rate of decrease in pH value slowed as the volume increased. The minimum value recorded was approximately 5 at 3500 L.



**Figure 2.** Change in the pH value of wastewater

##### 3.2.2. Alkalinity

Alkalinity is a measure of the water's ability to neutralize acids and resist changes that cause acidity, thus keeping its pH value constant. Titration is used to measure the alkalinity of a water sample, and water with a pH value of 8 or higher is considered alkaline. The pH scale is used to measure the alkalinity of water, with a pH between 8 and 10 being slightly alkaline and a pH of 10 or higher being very alkaline. A pH of 7 is considered neutral, while a pH less than 7 is considered acidic. Because HCO<sub>3</sub><sup>-</sup> is formed as a result of the reaction and carbonic acid is an aqueous solution of carbon dioxide, it has acidic properties. Therefore pH and alkalinity decreased. Since the carbon content of the water increased, COD increased.

In Figure 3, the alkalinity values of the wastewater are shown at a constant temperature of 20°C and a volume range of 0 to 4000 liters. After examining the graph, it is evident that the highest alkalinity value occurs at 700 liters, at approximately 330. This value then decreases until reaching around 2500 liters, where it drops to approximately 140. Afterwards, there is a lower rate of decrease as the volume increases, with the lowest observed value being 130 at 3500 liters.

### 3.2.3. COD

COD refers to the amount of oxygen consumed by oxidizing the raw material under heating conditions with chemical oxidants, usually expressed in terms of oxygen consumption in  $\text{mg L}^{-1}$  or  $\text{g L}^{-1}$ .

Figure 3 shows the COD values of wastewater at a constant temperature of  $20\text{ }^{\circ}\text{C}$ , a constant pressure of 10 bar, and a constant flow rate of  $10\text{ L min}^{-1}$ . Looking at the graph, it can be seen that the COD value in the water increases as the volume increases, starting from approximately  $40\text{ mg L}^{-1}$ . The lowest COD value was found to be approximately  $40\text{ mg L}^{-1}$  at 500 L, while the highest COD value was approximately  $80\text{ mg L}^{-1}$  at approximately 3300 L. Additionally, it can be observed from Figures 3 that when the amount of biogas supplied to the 300 L constant liquid absorption system with a constant flow rate of  $10\text{ L min}^{-1}$  reaches 2500 L, the water in the tank becomes saturated and the purification efficiency starts to decrease. These graphs demonstrate that 300 L of water can purify 2500 L of raw biogas at a constant pressure of 10 bar.

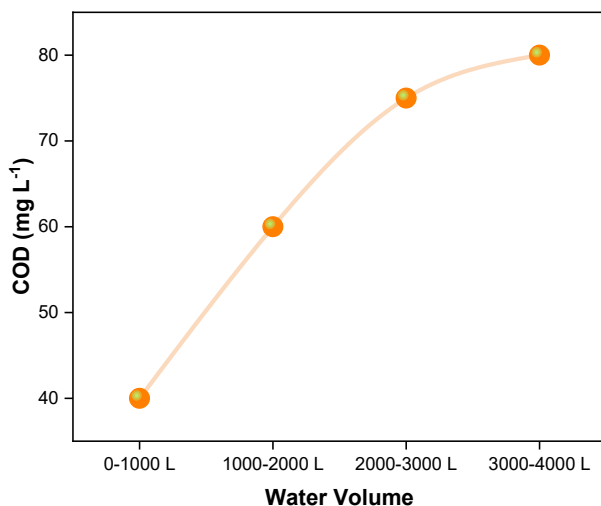


Figure 3. Change in the COD value of wastewater

## 4. Conclusion

In this study, water scrubbing method, which is one of the most effective methods among biogas purification or upgrading technologies, was applied. The factors affecting the efficient removal of  $\text{CO}_2$  from biogas and the production of biomethane ( $\text{CH}_4$ ) were extensively investigated. In the washing tower prepared for  $\text{CO}_2$  removal efficiency from biogas, it was observed that the absorption efficiency of  $\text{CO}_2$  decreased (from 37% to 3%) as the pressure of biogas supplied at constant flow rate increased from about 3 bar to 8 bar. The pH values of the wastewater passed through the tower (in the range of 0 to 4000 L) were measured to be approximately 7.7 at a volume of 500 L and approximately 5 at 3500 L. The alkalinity values of wastewater were determined between 330 and 140 values for low and high-water volume. The COD value in the water was approximately  $40\text{ mg L}^{-1}$  in 500 L. The highest COD value was 80 at approximately 3300 L. The results of this study show that  $\text{CO}_2$  removal from the biogas content using wastewater in the water scrubbing method is quite successful. In addition, the fact that the

biogas used in the study was produced in a real plant added a lot of originality to the study.

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## Disclosure statement

The author(s) declare no potential conflicts of interest.

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