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Removal of Hydrogen Sulfide from Biogas by Using the Water Scrubbing Techniques

Hossein Noruzi Moghadam[a,b] , Aghdas Banaeia [a,] *

[a] *Research Institute of Applied Science, Academic Center of Education, Culture and Research (ACECR), Tehran, Iran*

[b] *Razi Vaccine and Serum Research Institute, Agricultural Research, Education and Extension Organization (AREEO) Tehran, Iran*

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ABSTRACT

Renewable energy is one of the most important sources of energy supply in the world. Biomass power plants produce biogas under anaerobic conditions by burying organic waste. Biogas is being produced in the Mashhad Recycling Industrial Complex. This fuel is for consumers in the biogas plant to generate electricity. In this study, we performed the removal of hydrogen sulfide using water-scrubbing techniques in the form of gravity and stairs from biogas. The results show that the removal value of hydrogen sulfide varied between 34 to 87 and the removal efficiency of hydrogen sulfide varied between 34 to 97%. In addition, the loading rate of hydrogen sulfide varied between 8.1 to 40.8 $g/m^3.h$. Moreover, the elimination capacity of hydrogen sulfide varied between 7.3 to 20.9 g/m^3. h. This investigation is a novelty for design and construction in water Scrubbing. Water scrubbing systems compared to similar examples have distinguishing features in the vertical and stepped arrangement and suction to separate the remaining gases by a vacuum pump. The most important feature is the gravitational force-displacement of the water pump to flow water. Therefore, fewer water pumps are used. In this case, energy consumption is reduced.

1. Introduction

1.1. Biogas properties

The biological digestion of organic matter under anaerobic conditions produces biogas [1, 2]. Depending on the chemical composition of the material, and the time and conditions of anaerobic digestion, the biogas produced will vary in terms of the percentage of gaseous composition [2-4]. Biogas contains methane, carbon dioxide, hydrogen sulfide, nitrogen, hydrogen, oxygen, ammonia, and a small number of other elements so various scientific articles have reported the amount of methane about 40-75%, the amount of carbon dioxide to be about 25-50% and the amount of hydrogen sulfide between 0-6 percent. [1-6]. Methane is the main substance in biogas, which is known as a source of clean new energy [1]. Hydrogen sulfide in biogas causes corrosion and wear in power generators, pumps, compressors, storage tanks, control valves, connecting pipes, and concrete and steel [1, 3, 7, 8]. Hydrogen sulfide biogas has adverse and harmful effects on the environment and health [9]. In combination with oxygen in the air, it produces sulfuric acid. Sulfuric acid reacts with water and causes corrosion [8]. Hydrogen sulfide produces sulfur dioxide by combustion [8, 9]. Sulfur dioxide has harmful effects on the environment [9]. Sulfur reacts with air vapor to cause acid rain [1]. In addition, hydrogen sulfide at 500 ppm can cause lung damage and death [10]. The presence of carbon dioxide and hydrogen sulfide is undesirable in many processes and currents. It is therefore important to remove them from the gas [11]. Various methods are used to separate the gas mixture. One of the most important methods is absorbed in the liquids [12]. One of the new methods of gas separation is the formation of gaseous hydrates. Hydrated gas crystals are made from host gas molecules and water [13]. Remove impurities from biogas with pressurized water using a biogas treatment method. In this method, pollutant compounds could be physically adsorbed or dissolved in a liquid solution. [14]. Biogas is similar to natural gas in physical and chemical properties [1].

* Corresponding author.; e-mail: banaei@acecr.ac.ir

1.2. Gaseous hydrate structure

One of the new methods of gas separation is the formation of gaseous hydrates [13]. The general formula of gas hydrates is MnH2O, which M indicates the number of gas molecules by studying the structure of the water molecule, the researchers found that hydrogen bonds stabilize the structure of water by forming a ring of the form [15].In this case, there are voids in the structure of water at close distances. If these voids are filled with gas molecules, they form crystallized hydrated gas molecules, even at temperatures above the freezing point. Gas hydrate has an ice-like structure that usually forms at high pressure and low temperature and each gas molecule is placed in a cage of water molecules in each of its constituent units [16]. These units have two types of hydrated gas structures called hydrated gas structures 1 and 2. The gas hydrate equilibrium phase is mainly related to the ratio of gas molecules inside the hydrate cage [17].

1.3. Kinetics of gas hydrate formation

The kinetics of gas hydrate formation has three stages. These three steps include dissolution, nucleation, and growth. The first step is to dissolve the gas in the liquid (water) phase. This step continues continuously until the liquid is saturated. To reach the nucleation stage, the liquid must reach supersaturating. In the supersaturated state, a large amount of gas in the liquid phase is dissolved more than normal under the same conditions of temperature and pressure. The growth phase begins when the hydrate gas clusters are large enough to form a single nucleus [18]. In other words, these three stages include the penetration of gas into the water surface, surface migration and the formation of primary nuclei, and the growth stage of crystals [12].

1.4. Municipal wastes

Every year, Mashhad Municipality buries thousands of tons of municipal waste in two landfill sites. One of the landfill sites is the Torogh recycling industrial complex, which produces biogas from buried waste in anaerobic conditions. Biogas is produced at a recycling industrial complex at 400 m3/h. This fuel is consumed in three generators, while 620 k w electricity is produced. The Mashhad municipal waste management organization and Research Institute Applied Sciences (Shahid Beheshti University Tehran, Iran) have signed a contract. This is a compromised study, design, construction, operation, and delivery of a pilot unit for removing hydrogen sulfide from biogas. We perform water-scrubbing techniques in gravity and stairs for purifying five cubic meters of biogas per hour.

2. Materials and methods

2.1. The Components of Gravity and Stair System in Water Scrubbing Technique

Stair structure is the design and manufacture of iron. The stair has three meters in height and consisted of four steps to deploy the equipment. The height of the first stair is one meter, and the others have 70 cm. The equipment and tanks are placed on the platform with no vibration throughout the process (Figure 1). The steel water scrubber tower is designed and manufactured with a height of 70 and a diameter of 10 cm. The water scrubber tower is open and close for the necessary controls from three points. In the water scrubbing reaction biogas elements and water molecules, occur in stainless steel wires inside the tower. Methane in biogas has a lower solubility in water than carbon dioxide and hydrogen sulfide; it exited from the top of the water scrubber tower. Water scrub carbon dioxide and hydrogen sulfide from biogas. Biogas by a compressor (TEHRAN SANAT, 2HP, 100 L) and water by a water pump (LEO-LVR2-12) with a pressure of 2-10 bars are pumped into the water scrubber tower. Water and biogas flows were counter-currants thus water enters from the top and exited from the bottom, while the raw biogas entered the bottom and exited from the top of the tower (Figure 2). The primary moisture absorber is placed inside the water scrubber tower above the water inlet. A cylindrical column is placed inside the water scrubber tower under the water inlet. In this method, water flows into several tanks due to gravity. The volume of each tank is 100 l. After the water scrubber tower, the rest tank is found on the fourth step platform. The water exits the scrubber tower with carbon dioxide and hydrogen sulfide entering the rest tank through a pipe. In the water scrubbing method, hydrate formation occurs during the separation phase. In the resting tank, the two gases form hydrates in the water. In the rest tank, the water pressure reached two bars. In addition, the rest tank had a water outlet to the stripper tank and an emergency water outlet attached to the main tank (storage tank). Water from the rest tank entered the stripper tank and this tank is placed on the third platform. The stripper tank is equipped with an electric motor (TEC 0.25 KW) and gearbox (NMRW 040) that rotated a stainless steel propeller in 28 rounds per minute. An air blower (PALMA PG370) attaches the stripper tank with a control hose. When the scrubber system is switched on, the blower was air-pumped into the tank. Water from the stripper tank entered the vacuum tank. The vacuum tank is placed on the second platform and it is equipped with an oil, electric vacuum pump (FY 2B), which attaches to the tank with a control valve hose. This vacuum pump is a permanent vacuum of the residual gases from the water. The water output of the vacuum tank entered the sedimentation tank. The sulfur particles that form through the reaction in the stripper tank are precipitated in the sedimentation tank. Finally, water entered the storage tank from the sedimentation tank. The storage tank is placed on the ground the volume of the storage tank is 1000 l. The storage tank is equipped with a freshwater inlet pipe and buoyancy is attached to the pipe. This is found on the surface of the water to control the flow of water. This is a semi-closed system. When the scrubber system is switched on, the water pump is pumping pressurized water into the scrubber tower from the storage tank. In addition, the water output of the storage tank after flowing into the system returns to the storage tank (Figure 3).

Figure 1. Stair and Tank

Figure 2. Water scrubber tower

Figure 3. A view from the pilot plan of water scrubbing techniques

2.2. Analysis

For the biogas analysis, a control valve and a connector hose equipped with a pneumatic valve are applied, and raw biogas entered the compressor. Measure various elements in the raw biogas is performed by using the Geo-Tech Analyzer device (GA 2000) and a gas dryer is installed on the system. The control equipment included water flow meters and biogas flow meters, and the main distribution panel of the electrical circuit and biogas burner is installed on the system. In the designed system, hydrogen sulfide is converted into sulfur in a reaction with oxygen. Following that, the sulfur produced in the stripper tank is precipitated in the sedimentation tank [19].

$$2H_2S + O_2 \rightarrow 2S + 2H_2O$$

The experimental operation of the system is applying for 16 days (January 6-22, 2018). The difference between the initial input and the output value of hydrogen sulfide was calculated which was called the removal value (RV). The removal value (RV) system is calculated based on data sample from H_2S in a row and pure biogas by using Equation 1:

$$\mathrm{RV[ppm]} = \mathrm{H_2S_{input}[ppm]} - \mathrm{H_2S_{output}[ppm]} \quad (1)$$

The removal efficiency (RE) system is calculated based on data samples from H_2S in a row and pure biogas by using Equation 2 [20].

$$RE = \frac{H_2S_{input}[ppm] - H_2S_{output}[ppm]}{H_2S_{input}[ppm]} \times 100 \quad (2)$$

The amount of the hydrogen sulfide loaded rate (LR) in the system is a function of the inlet gas flow and concentration of the hydrogen sulfide inlet and molecular mass, along with the function of the filter volume and molecular volume of the gas at standard conditions. Therefore, the LR is calculated by using Equation 3 [20].

$$LR = \frac{gase\ flow\left[\frac{l}{h}\right] H_2S_{input}[ppm]34.08[\frac{g}{mol}]}{filter\ volum[m^3]22.41[\frac{l}{mol}]10^6} \quad (3)$$

The elimination capacity of hydrogen sulfide (EC) in the system is a function of the inlet gas flow and the difference between the concentration of the hydrogen sulfide inlet and outlet and molecular mass, along with the function of the filter volume and molecular volume of the gas at standard conditions. Therefore, the EC is calculated by using Equation 4 [20].

$$EC = \frac{gas\ flow\left[\frac{l}{h}\right]\{H_2S_{input}[ppm] - H_2S_{output}[ppm])34.08[\frac{g}{mol}]}{filter\ volume[m^3]22.41[\frac{l}{mol}] \times 10^6} \quad (4)$$

3. Results and Discussion

Figure 4 shows a schematic map of the components of gravity and stair system using the water scrubbing techniques. In addition, based on equation 1,2,3,4 the RV, RE, LR, and EC have been calculated respectively. The results based on Table 1 show that the biogas flow rate varied between 2.1 to 7.3 l/h. The mean of the biogas flow rate was 4.2 l/h. Table 1 shows the removed value (RV) of hydrogen sulfide sample analysis of the system. The results based on Table 1 show that the removal value of hydrogen sulfide varied from 34 to 87 and the mean of removing the value of hydrogen sulfide was 53. Figures 5-7 show a schematic diagram that results in the RE and LR, EC of hydrogen sulfide in the system. The results, based on Figure 5 show that the RE percentage of hydrogen sulfide varied from 34 to 97. The results based on figure 6 show that the LR of hydrogen sulfide varied between 8.1 to 40.8 g/m^3h. The results based on Figure 7 show that the EC of hydrogen sulfide varied between 7.3 to 20.9 g/m^3h. These changes in results depend on the temperature, flow rate, pressure, and freshness of the water. In addition, flow, pressure, and initial concentration of hydrogen sulfide in biogas are effective on the RV results. Freshwater is more capable of removing large amounts of hydrogen sulfide than regeneration water that is used continuously. The replacement of the stainless steel wires inside the water scrubber tower is effective on the RE rate. The new stainless steel wires have more removal compared to the old ones.

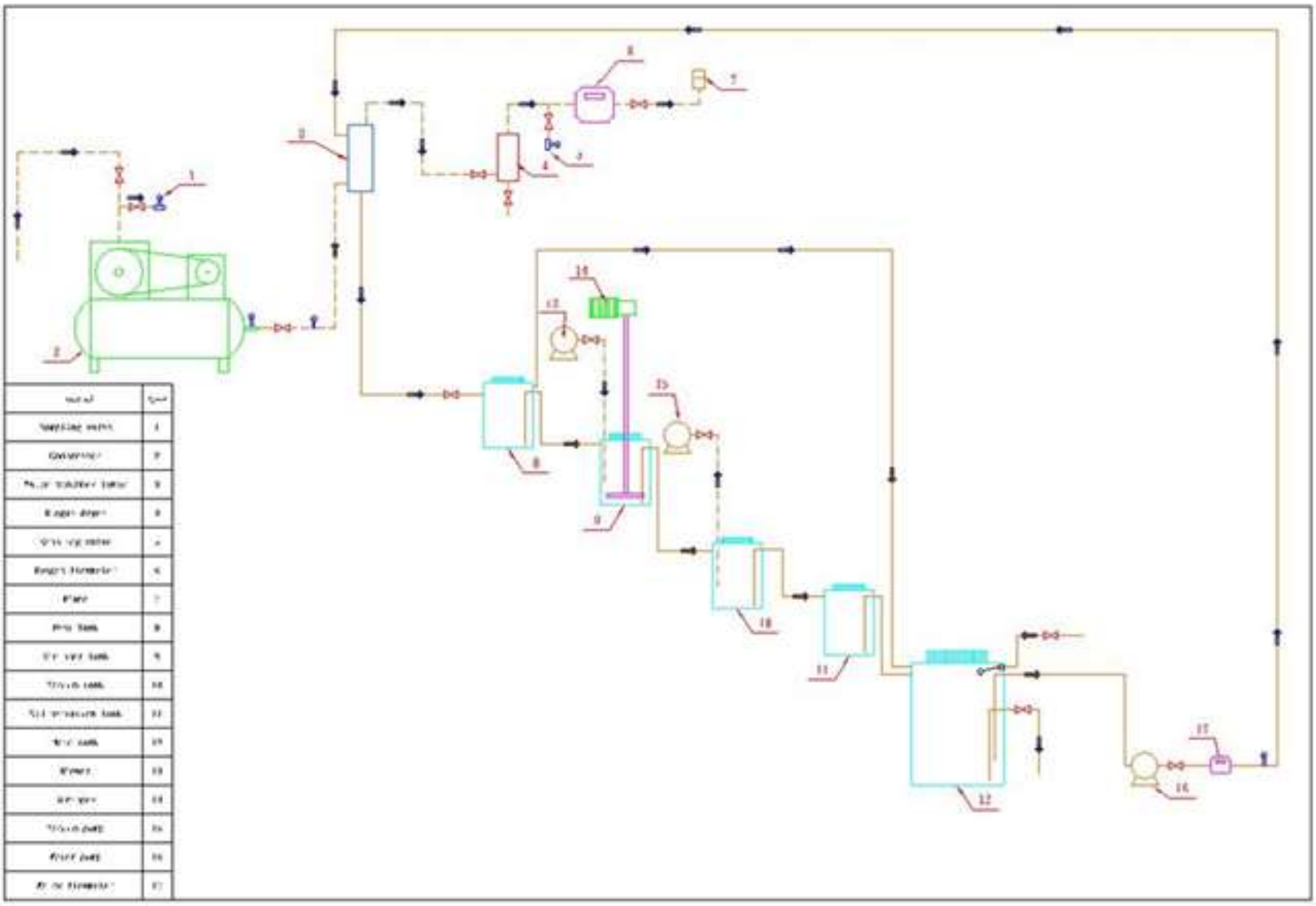

Figure 4. Schematic Diagram of Hydrogen Sulfide Removal from Biogas

Table 1. Sample Analysis of removed value (RV) of Hydrogen Sulfide in System

Analysis No.	H_2S Input (ppm)	H_2S Output (ppm)	H_2S Removed value(RV) (ppm)	Biogas Flow (l/h)	Biogas Flow* (m^3/h)
1	57	23	34	4.8	672
2	60	14	46	4.3	602
3	75	14	61	3.6	504
4	75	14	61	3.1	434
5	97	10	87	3	420
6	71	15	56	2.3	322
7	81	18	63	5.8	809
8	66	10	56	2.7	378
9	71	2	69	2.1	294
10	71	10	61	2.7	378
11	89	59	30	6.3	882
12	77	11	66	5.8	812
13	84	40	44	4.8	672
14	95	18	77	5	700
15	97	19	78	3.8	532
16	98	20	78	4.5	630
17	88	36	52	4.5	630
18	97	41	56	4.2	588
19	101	46	55	4.1	574
20	99	46	53	4.2	588
21	78	43	35	6.7	938
22	84	45	39	5.1	714
23	91	42	49	4	560
24	92	52	40	3.6	504
25	86	45	41	4.4	616
26	87	48	39	3.2	448
27	81	43	38	3.7	518
28	86	43	43	4	560
29	103	61	42	7.3	1022
30	81	40	41	3.3	462
Mean	83.9	30.9	53	4.2	592.1

**1 m^3 biogas equal to 140 liters in the system.*

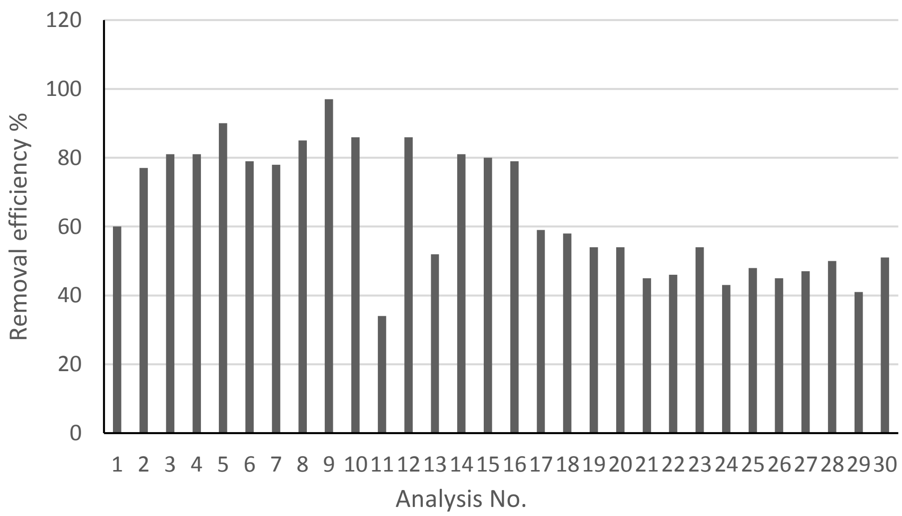

Figure 5. Results of Removal efficiency (RE) of Hydrogen Sulfide

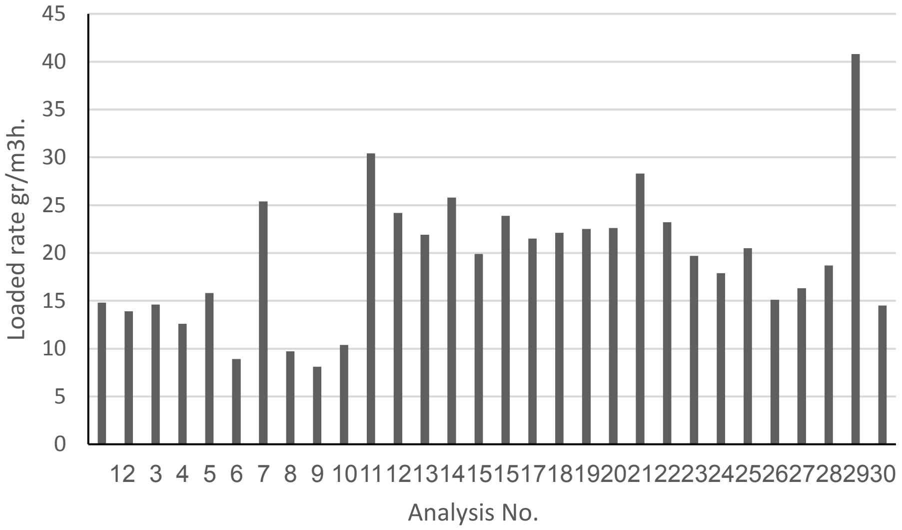

Figure 6. Results of loaded rate (LR) of Hydrogen Sulfide

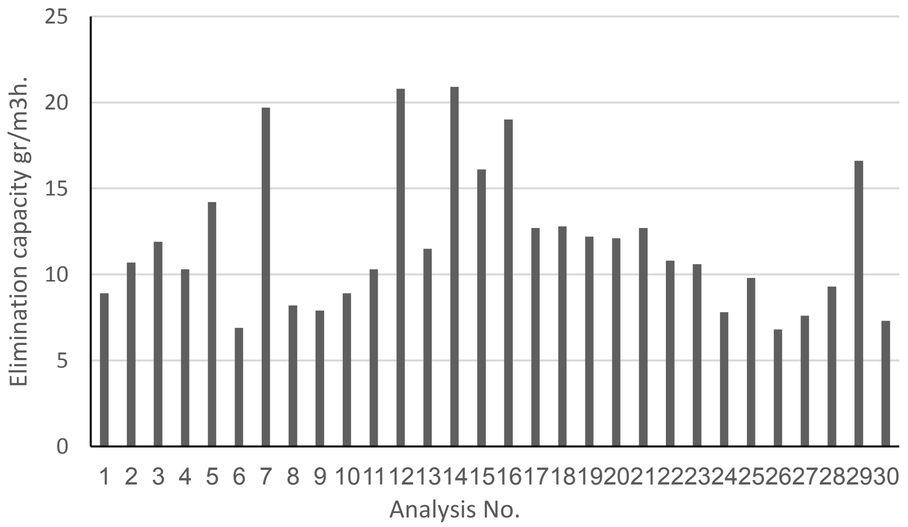

Figure 7. Results of Elimination capacity (EC) of Hydrogen Sulfide

This investigation is a novelty for design and construction in water Scrubbing. Water scrubbing systems compared to similar examples have distinguishing features in the vertical and stepped arrangement and suction to separate the remaining gases by a vacuum pump. The most important feature is the gravitational force-displacement of the water pump to flow water. Therefore, fewer water pumps are used compared to other water scrubbing techniques; in this case, energy consumption is reduced compared to other water scrubbing techniques. In the water scrubbing technique, compressed biogas is imported from the bottom and compressed water is imported from the top of the scrubber tower and inside the scrubber is a column of packed material [7, 21, 22]. The packed column increases the contact surface for absorption [22]. Water Scrubbing is robust [23], widely used and economical [10], commercially accessible [1, 24], simple, cheapest technology [21, 24], and environmentally friendly [5, 10]. Henry's constant indirectly expresses the solubility of a gas in a liquid. Temperature, pressure, and the type of adsorbent system affect solubility [25] affect solubility. As the temperature increases, the solubility of the gas in the liquid decreases, so the dissolution rate of carbon dioxide and hydrogen sulfide increases with decreasing water temperature, and vice versa [2, 25, 26]. Carbon dioxide and hydrogen sulfide are physically dissolved in high-pressure water [27]. On the other hand, at a high volumetric biogas rate, the contact time between water and gas molecules is reduced so the ability of water to absorb gas is reduced. At slow volumetric rates, the contact time between water and gas molecules increases, so more molecules are absorbed by the water. When the volumetric flow of water is high, more absorption is done [9]. Two methods are used for water scrubbing, including water scrubbing with regeneration and water scrubbing without regeneration. Water scrubbing with regeneration requires lower levels of water solvent and higher energy consumption [27]. In water scrubbing, the removal efficiency of hydrogen sulfide increases with the increasing water level, which means that its biogas concentration decreases, and the higher the gas flow, the lower its concentration in biogas. On the other hand, with increasing time, the removal efficiency decreases. Therefore, the concentration of hydrogen sulfide in biogas increases [5, 28]. In this case, it is necessary to replace or regenerate water [28].Olugasa et al. investigated the design and construction of a water scrubber for the upgrading of biogas. The results showed that the H_2S concentration was reduced from 1% to 0.4%. [29]. Based on the result the RE was 60%. Tira et al. tested the improving biogas quality through the circulated water scrubbing method. The results showed that RE was 59.7% [9].Lien et al. investigated the water scrubbing for removal of hydrogen sulfide (H_2S) in biogas from Hog Farms. The results showed that the RE was 78.3 and 30.6% in water scrubbing times 30 and 90 seconds respectively [28]. Ou et al. Performed research on the removal of hydrogen sulfide from biogas using a bubbling tank fed with aerated wastewater. The results showed that the H_2S concentration was 907 ± 212 ppm. The results showed that the RE was 86 to 71 percent [8]. These results compare to the results obtained in the present study with a mean of RE was 53% they adapt according to different execution conditions. Cuimei et al. investigated the dynamic control design and simulation of the biogas-pressurized water scrubbing process[30]. The base of this process for the removal of CO2 was similar to the present study for the removal of H_2S while it varied in performance. Mamun et al. Evaluated the removal of hydrogen sulfide from biogas using Zerovalent iron, and the mean H_2S concentration of 211, 138, and 139 ppm are introduced into the chemical H_2S elimination system, the H_2S concentration could reach below 50 ppm [31]. Based on this result the RE was 74 and 60%. In this research, the process of water solution was similar to the present study process, while it differed in the absorption reaction method. Thus compared to the results obtained in the present study RE was more than in our studies. Farooq et al. At the pilot plant of biogas upgrading, using various absorbents, the H_2S concentration after the chemical absorption from 100 ppm decreased to 40-45 ppm [32]. Based on the result the RE was 60 and 55%, while it varied in chemical absorption, method thus comparable to the results obtained in the present study RE was similar to our studies. Pinate et al. Investigated the removal of hydrogen sulfide (H_2S) from biogas the results indicated that the absorbents could reduce the H_2S from 3,141 to 0 on the first day and 6 ppm on the 25th day [33]. Based on the result the RE was 100 and 99.8% in the present study, the RE was lesser than the mentioned research. Vrbová et al. upgrading biogas to biomethane using a membrane. Pilot-scale results indicated that the H_2S concentration of 100mg/m^3 biogas a flow rate equal to7 m^3/h reach 21.25 mg/m^3 [34, 35]. Based on this result the RE was 79 %, which was similar to the maximum RE reported in the present study. The amount of the produced methane along with hydrogen sulfide is associated with the materials contained and the population of the bacteria in landfills. In addition, the most influential factor is a high amount of H_2S in anaerobic digesters in biogas production, and primary sludge and solid loading rates are infecting H_2S production [36-40]. Hydrogen sulfide production correlated with sulfate reduction and sulfate-reducing bacterial activity in anaerobic environments [41-45]. Skerman et al. Investigated the practical options for biogas cleaning before on-farm use at piggeries, and the H_2S concentration was reduced from 4,000 to lesser than 400 ppm. Results showed that the RE of H_2S from raw biogas was more than 90% [46-50]. In the present study, the RE was lesser than the mentioned research. Process of methane production in normal anaerobic conditions, higher amounts of hydrogen sulfide in methane are considered undesirable. Therefore, if the amount of protein with cysteine and cysteine amino acids in the buried waste is higher, the amount of the produced hydrogen sulfide increases.

4. Conclusion

Hydrogen sulfide in biogas is produced in large quantities. This amount of hydrogen sulfide can destroy power generators. The results of this research showed that this water scrubbing technique has been able to remove up to 97% of hydrogen sulfide on average, regardless of the amount of hydrogen sulfide in the gas flow. Consumable sulfur is extracted from the removed hydrogen sulfide. These water-scrubbing systems compared to similar examples have distinguishing features in the vertical and stepped arrangement and suction to separate the remaining gases by a vacuum pump. The most important feature is the gravitational force-displacement of the water pump to flow water. This system will be able to remove hydrogen sulfide all the time in 70-97% with a few changes in the height of the tower and packing the bed. A complementary technique, such as solid-state filters, can use to complete and optimize removal conditions.

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