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Improving The Removal Efficiency of Carbon Dioxide and Hydrogen Sulfide from Biogas Using Different Stirring Methods for Chemical Solutions Used in The Purification Process

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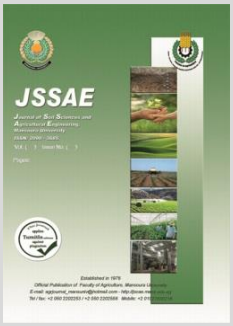

ABSTRACT

The main purpose of this study was to improve the biogas quality by keeping the methane content above 90%. To achieve this goal, it has been used a combined stage of slaked lime solution 50%and sodium hydroxide of 10 and 20% respectively at different stirring methods. The stirring methods were; without stirring, manual stirring, and mechanical stirring at speeds of 250, 200 and 150 rpm.The obtained results indicated that, a combined stage of slaked lime solution 50% and sodium hydroxide of 10%and 20%respectively with mechanical stirring at speed of 150 rpm showed a highly removal performance compared with the other different treatments. The methane content was above 99% through the first 32 hours compared with 19, 28 and 29 hours for manual stirring and mechanical stirring speeds of 250 and 200 rpm respectively. Moreover, the methane content was remained above 90% at removal time of 44 hours compared with 32, 40 and 41 hours for manual stirring and mechanical stirring at the same speeds respectively. In addition, the mechanical stirring at speed of 150 rpm gave the highest average removal efficiency of carbon dioxide 95.9% which keeping it less than 10% compared with 92.1, 94.6 and 94.7% for manual stirring and mechanical stirring at speeds of 250 and 200 rpm respectively. On the other hand, the highest average removal efficiency of hydrogen sulfide during purification process period off 44 hours was 79.2% compared with 78.5 and 78.9% for the two other mechanical stirring speeds of 250 and 200 rpm respectively.

Keywords: Mechanical Stirring - Biogas Purification - Co2 Capture - Chemical Scrubbing

INTRODUCTION

There are concerns about the excessive use of fossil fuels due to its harmful impact on the environment, especially the emission of carbon dioxide that is considered a major reason of greenhouse gas (GHG) (Dennis Y.C. *et. al*, 2014). As a result, there is interest from researchers and environmentalists about methane fuel production from biogas via anaerobic digestion (AD). Biogas with enriched methane is an important renewable fuel; it is environmentally friendly, clean, cheap and versatile (Vijay, *et.al*, 2012).

Biogas can be used directly to generate power, but the large volume of carbon dioxide reduces the caloric value of the gas, increasing compression and transportation costs and limiting economic feasibility to uses that occur at the point of production. Purification allows for a wider variety of uses, either for heat and electricity, or for vehicle fuels. For use as a fuel, purification to remove carbon dioxide and hydrogen sulfide is required, because hydrogen sulfide corrodes vital mechanical components within engine generator sets and vehicle engines if it is not removed (Zhao, *et. al*., 2010). Two major steps are performed for biogas purification process: (1) A cleaning process to remove the trace components and (2) An upgrading process to adjust the calorific value. Upgrading is generally performed in order to meet the standards for use as vehicle fuel or for injection in the natural gas grid (Ryckebosch, *et.al*, 2011).

Chemical scrubbing can produce gas with a high level of methane content of more than 95% and no methane loss, hence it's widely used. The chemical used can either be caustic solvent, amines solvent, amino acid salt (Maile *et al*., 2017). One of the most important steps in the biogas upgrading process is the selection of solvent. Utilizing the solvent appropriateness for absorbing carbon dioxide from biogas is determined by the solubility differential of carbon dioxide and methane in that solvent. Carbon dioxide could be absorbed from biogas by utilizing water selectively in a pure physical process. Moreover, the solvent must comply with certain criteria including availability, low cost, be environmentally friendly, have a high carbon dioxide load, easy to regenerate, and low viscosity to achieve solubility gradient between carbon dioxide and methane (Wilken *et al*., 2017).

(Abdeen *et al*., 2016) said that the most abundant and widely used solvents are caustic solvents such as sodium hydroxide, potassium hydroxide, and calcium hydroxide which able to absorb carbon dioxide from the gases mixture after amine solutions. Several studies of sodium hydroxide have been considered as an effective carbon dioxide absorber. sodium hydroxide has several advantages such as lower cost, more abundant and possessing a greater theoretical carbon dioxide capture capacity. Carbon dioxide is chemically absorbed through the given reaction in Equation. (1):

$$2NaOH_{(aq)} + CO_{2(g)} \rightarrow Na_2CO_{3(aq)} + H_2O_{(l)} \quad (1)$$

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They also added that, the absorption of carbon dioxide using carbonate solutions has many advantages, such as the low volatility, low corrosion rate, and the possibility to absorb other impurity gases. Several early works discussed the use of carbonate solutions in absorbing and capturing carbon dioxide. When a carbonate solution is used to absorb carbon dioxide, the overall reaction can be written as Equation. (2).

$$CO_{2(g)} + CO_{3\,2-} + H_2O \rightleftharpoons 2HCO_3^- \qquad (2)$$

(El-Bakhshwan *et al*., 2015) reported that, the purification with sodium hydroxide at combined stages of 10 and 20% respectively was the best option for removing carbon dioxide and recommended further study for improving the biogas quality.

The main goal of this study was to improve the biogas quality by keeping the methane content above 90% and the carbon dioxide percentage less than 10%.

MATERIALS AND METHODS

Material

The experiment of work was implemented at the laboratory of Development of Biogas Production and Utilization System Project in the Tractors and Machinery Research and Test Station, Alexandria city - Agricultural Engineering Research Institute.

Biogas Composition

The biogas produced from two fixed dome digesters with total volume of 20 m3. It was collected and stored in a floating fiber glass gasholder with total volume of 20 m3. The biogas was passed in solution of ferric EDTA (Fe(III)EDTA) to reduce the hydrogen sulfide value before it has compressed. The biogas components are methane, carbon dioxide, and hydrogen sulfide. The average percentages of biogas contents after passed through ferric EDTA (Fe(III)EDTA) were; 64.4, 36.1, and 0.5% for methane, carbon dioxide, and oxygen respectively, while the average percentage of hydrogen sulfide was 248 ppm. These percentages of biogas components were precisely measured using a Portable biogas analyzer Model: GA5000 (Geotechnical (UK).

Biogas Scrubbing System

The main substrate used in this study was slaked lime (Ca(OH)2), it was used with a concentration of 50%. The second substrate was sodium hydroxide at different concentrations of 10 and 20% respectively. Combined stages of slaked lime solution at 50% and sodium hydroxide at concentrations of 10 and 20% respectively were used to remove the carbon dioxide and the hydrogen sulfide from biogas. Several bottles 5 liters volume were used in the purification process. These bottles were filled with slaked lime and sodium hydroxide and the biogas passed through these chemicals for the purification process.

Mechanical Stirring System

The mechanical stirring system was used to study the effect of stirring the slaked lime solution on the biogas purification process. The system is consisting of two units (stirring unit and speed control unit) as shown in Fig. (1). Fig. (2) shows a cross section area of the stirring unit components. Standard Frequency Inverter (CDI-EM60 Series Mini Type VFD) was used to control and adjust the speed of the stirring unit to ensure that the solution used in the purification process is not allowed to effervescence.

Methods

Hydrogen sulfide capture from biogas

The raw biogas was passed through the Ferric EDTA (Fe(III)EDTA) solution to remove hydrogen sulfide, then the biogas was compressed and used in the experimental work. Hydrogen sulfide removal efficiency was the ratio of the treated hydrogen sulfide and the inlet hydrogen sulfide concentration in biogas or absorbent solution as in Equation (3) (Rakmak *et al.* ,2010).

$$H_2S_{\,Renval} = \frac{[H_2S]\ inlet - [H_2S]\ outlet}{[H_2S]\ inlet}\ \% \qquad (3)$$

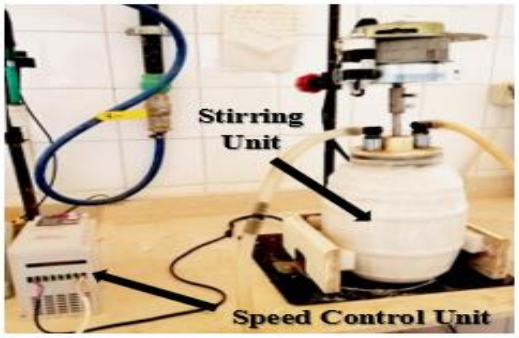

Fig. 1. Mechanical stirring system.

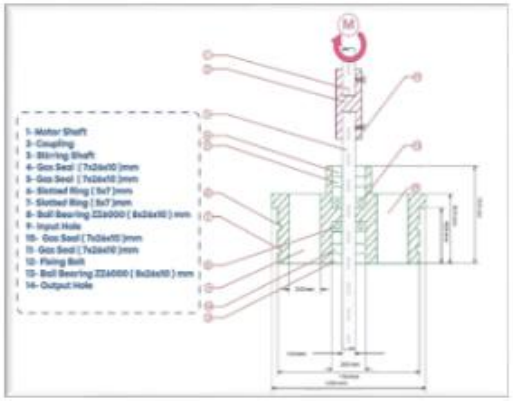

Fig .2. A cross section area of the stirring unit components.

Where % $H_2S_{\,Removal}$. is the hydrogen sulfide percentage removal efficiency, $(H_2S)_{inlet}$ is the inlet hydrogen sulfide concentration in biogas inlet, and $(H_2S)_{outlet}$ is the hydrogen sulfide concentration in biogas outlet.

Carbon dioxide capture from biogas

Using of sodium hydroxide at combined stages of (10 and 20%) was the best option for removing carbon dioxide (El-Bakhshwan *et al*., 2015). In this experiment, the same treatments have been used besides, slaked lime at 50% concentration as an initial filter in the purification process. The carbon dioxide removal efficiency was calculated according to the following Equation (4) (Maile *et al*. ,2017).

$$\eta_{Co_2} = \left(1 - \frac{Co_{2\,pur}}{Co_{2\,raw}}\right) x\ 100 \qquad (4)$$

Where: (η_{CO2}): carbon dioxide removal efficiency, (CO_{2pur}): carbon dioxide content of the purified sample, and (CO_{2raw}): carbon dioxide of the raw sample.

Stirring System:

Manual stirring has been done for the slaked lime solution during the purification process and the stirring period was 1 minutes every 20 minutes throughout the experiments time. Several experiments were conducted, to choose the proper speed to ensure the un-effervescence of the slaked lime solution during the removal process and the results showed that the maximum speed with no

effervescence was 250 rpm. So, others different speeds of 200 and 150 rpm were applied to identify the optimum stirring speed and its effect on the purification process.

RESULTS AND DISCUSSION

Effect of Different Stirring Methods on the Purification Process:

1- Effect of non-Stirring Method:

Two different concentrations of sodium hydroxide (10 and 20%) with the slaked lime of 50% without stirring in combined stages were used to remove carbon dioxide and hydrogen sulfate from biogas. The obtained results pointed in Fig. (3) showed that the carbon dioxide percentage reached 5.1% through the first nine hours, while it was reached a percentage of 10.2% after 12 hours. The purification efficiency of carbon dioxide ranged from 99.2% at the beginning of the experiment and to 85.9% with the average of 94.3% after 9 hours. Moreover, the carbon dioxide percentage reached 10.2% with purification efficiency of 71.7% after 12 hours of the purification process.

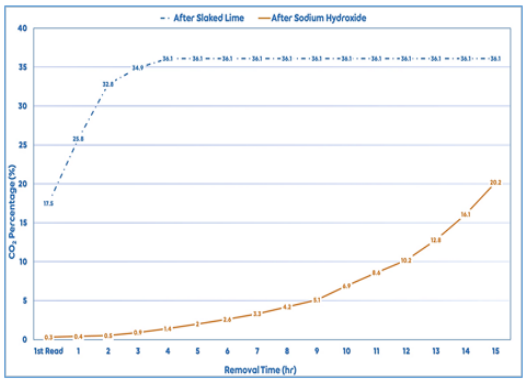

Fig. 3. Carbon dioxide percentage in biogas after the purification process using non stirring method.

In addition, the minimum value of hydrogen sulfide was 22 ppm at the first hour while the average removal efficiency through the first five hours was 90.3% and the maximum value was 99 ppm and it occurred after 15 hours of the process with a removal efficiency of 60.1%.

The methane content in the purified biogas was pointed in Fig. (4). The data clear that methane content was 94.9% with increasing ratio of 49.7% through the first nine hours. Moreover, the methane content approached 90% after 11 hours of purification. The average methane content through the first eleven hours was 96.9%.

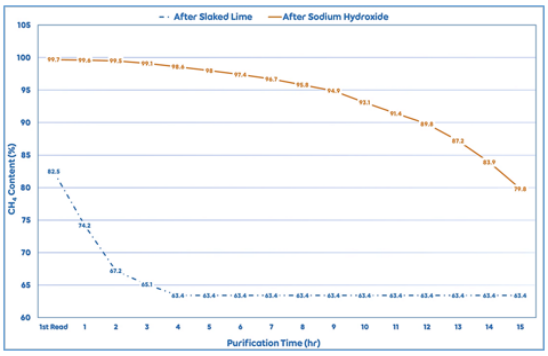

Fig. 4. Methane content in biogas after the purification process using non stirring method.

2- Effect of Manual Stirring Method:

Manual stirring of slaked lime solution was used in this phase with two different concentrations of sodium hydroxide (10 and 20%) to remove the carbon dioxide and hydrogen sulfide. The obtained data were pointed in Fig. (5). It was cleared that the carbon dioxide percentage remained less than 5% through the first 29 hours compared with 9 hours for the same treatment without stirring. Moreover, the carbon dioxide percentage remained less than 10% through 32 hour compared with 11 hours for the same treatment without stirring.

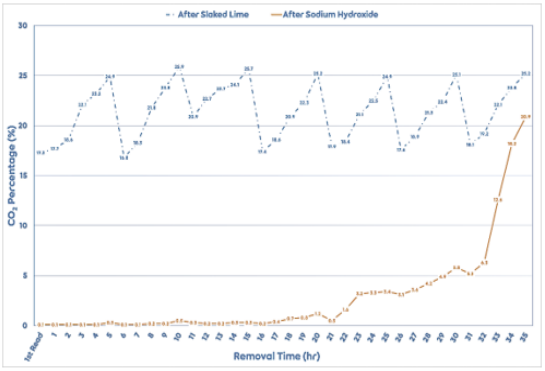

Fig. 5. Carbon dioxide percentage in biogas after purification process using manual stirring method.

The obtained results also, demonstrated that the manual stirring of slaked lime solution 50% with sodium hydroxide of 10% and 20% respectively enhanced the removal process of carbon dioxide. The removal efficiency of carbon dioxide ranged from 99.7% to 86.4% through the first 29 hours. The carbon dioxide percentage reached 6.3% at removal efficiency of 82.5% and removal time of 32 hours. On the other hand, the results cleared that exceeding the time of 32 hours, the carbon dioxide percentage was rapidly increased and reached 12.6% through only one hour. This mean that the optimum time for removing the carbon dioxide using the manual stirring of slaked lime solution 50% with sodium hydroxide of 10% and 20% respectively was 32 hours.

In addition, the hydrogen sulfide values were ranged from 15 to 53 ppm with average value of 33.9 ppm through the first 32 hours. The removal efficiency through this time was ranged from 94 to 78.6% with average of 86.3%. In all cases, the hydrogen sulfide was at the desirable limit.

On the other hand, using the manual stirring of slaked lime solution 50% with sodium hydroxide of 10% and 20% respectively gave the highest methane content compared with the non-stirring treatment. The data pointed in Fig. (6) showed that the methane content ranged from 99.9% to 93.7% through the first 32 hours of purification compared with only 10 hours for the non-stirred treatment. The obtained results also, indicated that the removal efficiency was higher and the removal time was increased with three time as compared with the same treatment without stirring.

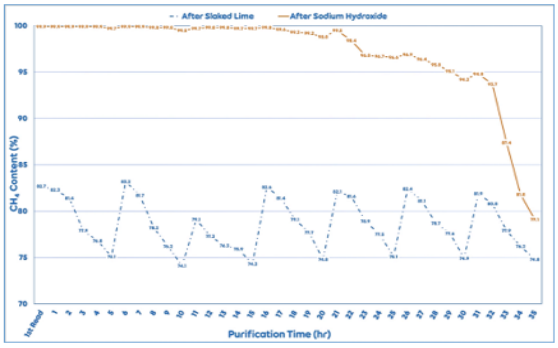

Fig. 6. Methane content in biogas after the purification process using manual stirring method.

3- Effect of Mechanical Stirring Method:

Effect of Mechanical Stirring at Speed of 250 rpm:

Mechanical stirring shows a great effect for removing both of carbon dioxide and hydrogen sulfide and increasing the removal time of slaked lime solution. The obtained results are illustrated in Fig. (7). The results show that, the carbon dioxide percentage reached 5% after 35 hours, while it was reached 10% after 40 hours compared with 32 hours with for manual stirring process.

The data also, revealed that, the removal efficiency of carbon dioxide ranged from 99.7% to 86.4% through the removal time of 35 hours. Meanwhile, the carbon dioxide percentage was less than 10% with removal efficiency of 73.1%, after 40 hours. In addition, the hydrogen sulfide value was 6 ppm at the first hour with a removal efficiency of 97.6%. While, the removal efficiency of hydrogen sulfide was decreased to 67.7% after 40 hours with a value of 80 ppm.

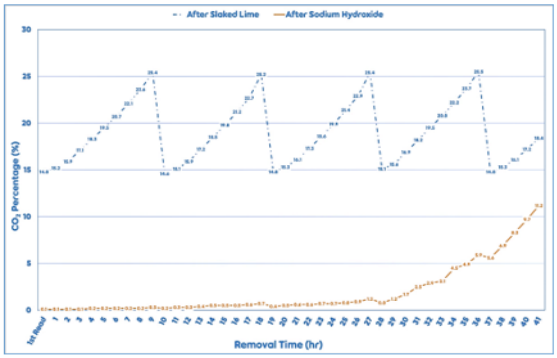

Fig. 7. Carbon dioxide percentage in biogas after purification process using mechanical stirring method at speed of 250 rpm.

The mechanical stirring showed a great effect on methane content during the purification process as shown in Fig. (8). The obtained results revealed that the slaked lime solution was stable for 9 hours which results in remaining the methane content above 75% compared with 5 hours for the manual stirring process.

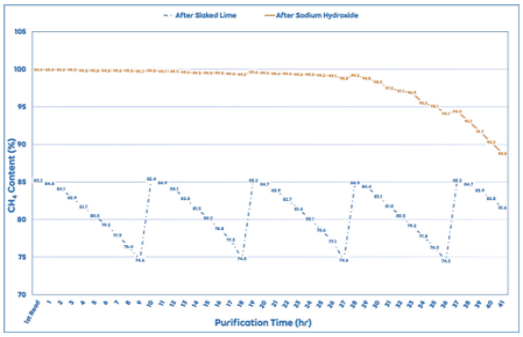

Fig. 8. Methane content in biogas after the purification process using mechanical stirring method at speed of 250 rpm.

The data indicated that the mechanical stirring of slaked lime solution increase its efficiency for removal carbon dioxide and hydrogen sulfide compared with manual stirring. On the other hand, the slaked lime solution was changed every 9 hours compared with 5 and 1 hours for manual and non-stirring methods respectively.

The methane content ranged from 99.9% at the first hour to 90.3% after 40 hours with average methane content of 98.3%. The increasing percentage ranged from 57.6 to 42.4% through the same purification time.

Effect of Mechanical Stirring at Speed of 200 rpm

The effect of mechanical stirring at speed of 200 rpm of slaked lime solution through the removal process was investigated and the obtained results are pointed in Fig. (9). The results show that, the carbon dioxide ranged from 0.1 to 5.1% through the first 37 hours. Moreover, it was reached 10% after 41 hours.

The data also, demonstrated that the removal efficiency for carbon dioxide ranged from 99.7 to 85.9% through the first 37 hours and it was decreased at a slight rate and reached 73.4% after 41 hours. At this efficiency, the carbon dioxide percentage was 9.6% (i.e. less than 10%).

On the other hand, the hydrogen sulfide ranged from 5 to 24 ppm with a removal efficiency ranged from 98.8% to 90.3% through the first 11 hours. The maximum value of hydrogen sulfide was 89 ppm with a removal efficiency of 64.1% after 41 hours. In general, the hydrogen sulfide was at the desirable and safety limit.

Fig. (10) shows the effect of mechanical stirring at speed of 200 rpm for slaked lime solution 50% with sodium hydroxide of 10% and 20% respectively on the methane content during the purification process. The data clear that the methane content was 99.9% at the first hour and it was slightly decreased to 90.4% after 41 hours compared with 90.3%, which occurred after 40 hours at speed of 250 rpm. The increasing ratio ranged from 57.6 to 42.6% at purification time of 41 hours. These results mean that the stirring speed of 200 rpm was higher efficiency than the stirring speed of 250 rpm. On the other hand, the slaked lime solution changed time was 9 hours of process (as the same of stirring speed of 250 rpm).

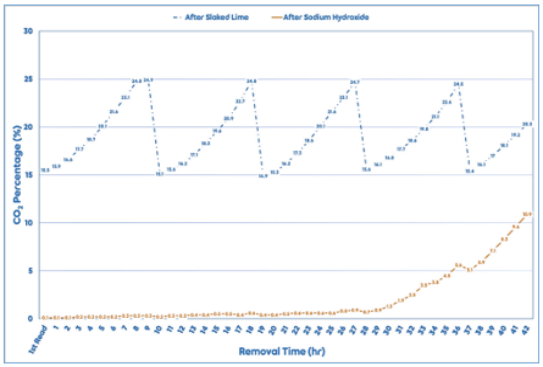

Fig. 9. Carbon dioxide percentage in biogas after purification process using mechanical stirring method at speed of 200 rpm.

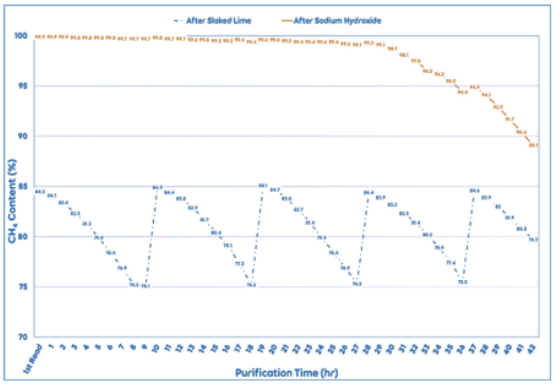

Fig. 10. Methane content in biogas after the purification process using mechanical stirring method at speed of 200 rpm.

Effect of Using Mechanical Stirring at Speed of 150 rpm

The effect of mechanical stirring of slaked lime solution at speed of 150 rpm on the removal efficiency was illustrated in Fig. (11). The carbon dioxide percentage was remained less than 1% for 32 hours compared with 28 and 29 hours for the two other stirring speeds of 250 and 200 rpm respectively. Meanwhile, the carbon dioxide percentage of 5.1% after 39 hours compared with 35 and 37 hours at stirring speeds of 250 and 200 rpm respectively. Moreover, the carbon dioxide percentage reached 10% after 45 hours compared with 40 and 42 hours for the other two stirring speeds of 250 and 200 rpm respectively.

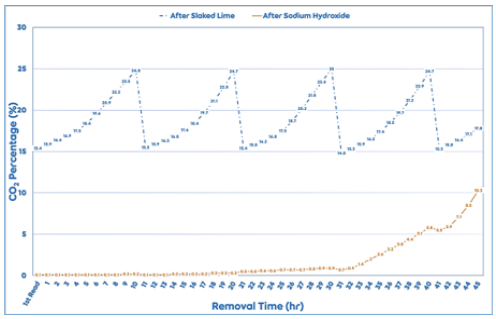

Fig. 11. Carbon dioxide percentage in biogas after purification process using mechanical stirring method at speed of 150 rpm.

The removal efficiency of carbon dioxide ranged from 99.7% to 97.5% with average of 99% through the first 32 hours of purification. While, the removal efficiency reached 85.9 and 71.5% with carbon dioxide percentage of 5.1 and 10.3% respectively at the exceeding time of purification. Moreover, minimum value of the hydrogen sulfide was 4 ppm at the first hour and reached the maximum value of 79 ppm after 45 hours with a removal efficiency ranged from 98.4 to 68.1%.

The mechanical stirring at speed of 150 rpm gave the highest methane content as shown in Fig. (12). The obtained results revealed that the methane content was above 99% through the first 32 hours compared with 28 and 29 hours for other two stirring speeds of 250 and 200 rpm respectively. In addition, the methane content was remained above 90% at removal time of 44 hours compared with 40 and 41 hours for the other two stirring speeds of 250 and 200 rpm respectively. On the other hand, using mechanical stirring of slaked lime solution 50% with sodium hydroxide of 10 and 20% respectively at speed of 150 rpm was the optimum treatment during the purification process.

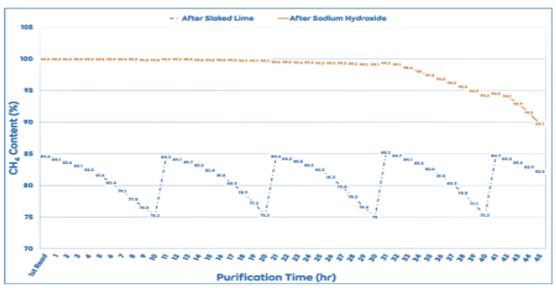

Fig. 12. Methane content in biogas after the purification process using mechanical stirring method at speed of 150 rpm.

The obtained results illustrated in Fig. (13) and Fig. (14) demonstrated that using mechanical stirring of slaked lime solution 50% with sodium hydroxide of 10 and 20% respectively at speed of 150 rpm showed a highly removal performance compared the other studied treatments as it keeps the methane content above 90% for 44 hours.

In addition, the mechanical stirring at 150 rpm gave the highest average removal efficiency of carbon dioxide during the purification process period. It was 95.9% compared with 92.1, 94.6 and 94.7% for manual stirring and the other two mechanical stirring speeds of 250 and 200 rpm respectively. Moreover, the methane content was above 90% for 44 hours compared with 32, 40 and 41 hours for manual stirring and the other two stirring speeds of 250 and 200 rpm respectively.

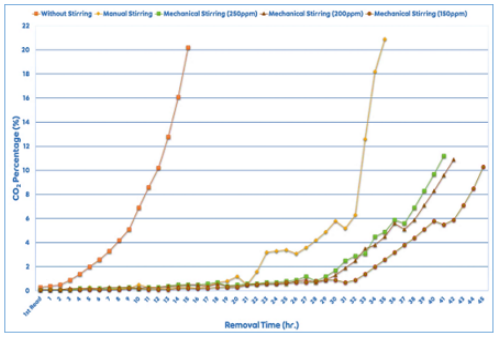

Fig. 13. The effect of different stirring methods on carbon dioxide percentage.

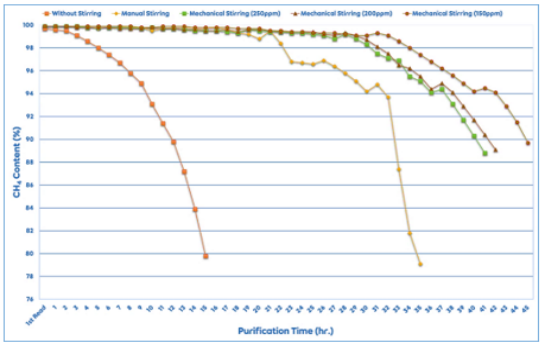

Fig. 14. The effect of different stirring methods on methane content.

On the other hand, the highest average removal efficiency of hydrogen sulfide during purification process using mechanical stirring 150 rpm was 79.2% compared with 78.5 and 78.9% for the two other stirring speeds of 250 and 200 rpm respectively. The obtained results illustrated in Fig. (15) demonstrated that the mechanical stirring at 150 rpm gave the best conditions for stirring of slaked lime solution which results in remaining of methane content stabled above 75%. The slaked lime changed time was 10 hours compared with 9 hours for the other two mechanical stirring speeds and 5 hours with for the manual stirring method.

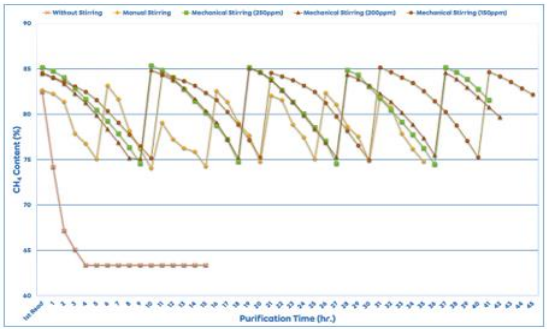

Fig. 15. Comparison of methane content at slaked lime using different stirring methods.

CONCLUSION

The obtained results of the present study concluded that the use of mechanical stirring of slaked lime solution 50% at speed of 150 rpm combined with sodium hydroxide of 10% and 20% respectively has a highly removal efficiency of carbon dioxide and hydrogen sulfide and consequently, higher methane content compared with the other different treatments. It was led to:

1. Maintain the methane content above 99% for 32 hours compared with 28 and 29 hours for the two other mechanical stirring speeds of 250 and 200 rpm respectively. While, it was not decreased to less than 90% for 44 hours compared with 32, 40 and 41 hours for manual stirring and the other two stirring speeds of 250 and 200 rpm respectively.
2. Increase the average removal efficiency of carbon dioxide during purification process to (95.9%) compared with 92.1, 94.6 and 94.7% for manual stirring and two other stirring speeds of 250 and 200 rpm respectively.
3. Increase the slaked lime solution changed time to 10 hours compared with 9 hours for the other two stirring speeds of 250 and 200 rpm and 5 hours for the manual stirring method.
4. The hydrogen sulfide was at the desirable and safety limits for different studied treatments.

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تحسين كفاءة عملية الإزالة لغاز ثاني أكسيد الكربون وكبريتيد الهيدروجين من الغاز الحيوي باستخدام طرق التقليب المختلفة للمحاليل الكيميائية المستخدمة في عملية التنقية.

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تعتبر تكنولوجيا الغاز الحيوي ذات أهمية كبيرة من خلال التنمية المستدامة مصر 2030، وتهدف تكنولوجيا الغاز الحيوي إلى إنتاج طاقة نظيفة جديدة ومتجددة وكبديل للطاقة التقليدية مع إنتاج سماد عضوي غنى بالعناصر الغذائية وخالي من مسببات الامراض مما يؤدي الى حماية البيئة من التلوث بالإضافة الي تحويل المتبقيات والمخلفات الزراعية الي مواد ذات مردود اقتصادي. يهدف البحث الي دراسة استخدام طرق التقليب المختلفة (بدون تقليب، تقليب يدوي، تقليب ميكانيكي بسرعات (150، 200 و250 لفة في الدقيقة)) على كفاءة المعالجة الكيميائية باستخدام محلول الجير المطفأ للتخلص من كلا من غاز ثاني أكسيد الكربون وكبريتيد الهيدروجين من الغاز الحيوي والوصول لمحتوي غاز الميثان لأكثر من 90%، وذلك باستخدام الجير المطفأ بتركيز 50% مع هيدروكسيد الصوديوم 10و20٪ على التوالي. اظهرت النتائج المتحصل عليها ان استخدام التقليب الميكانيكي عد سرعة 150 لفة/دقيقة لمحلول الجير المطفأ 50٪ مع هيدروكسيد الصوديوم 10٪ و20٪ على الترتيب هي أفضل معاملة أثناء عملية التنقية. وقد أعطي اعلى كفاءة إزالة مقارنة بالمعاملات الأخرى حيث حافظ على محتوى غاز الميثان أعلى من 90٪ لمدة 44 ساعة. بالإضافة إلى ذلك، كان متوسط كفاءة الإزالة لغاز ثاني أكسيد الكربون خلال عملية التنقية 95.4٪ مقارنة بـ 92.1 و94.6 و94.7 لكلا من التقليب اليدوي وسرعتي التقليب الأخريين 250 و200 لفة/ الدقيقة على الترتيب. وكان الوقت اللازم للحفاظ على نسبة غاز الميثان اعلي من 90% خلال عملية التنقية 44 ساعة مقارنة بـ 32 و40 و41 لكلا من التقليب اليدوي وسرعتي التقليب الميكانيكي 250 و200 لفة/ الدقيقة على الترتيب. من ناحية اخري، كان متوسط كفاءة الإزالة لكبريتيد الهيدروجين خلال عملية التنقية 79.2٪ مقارنة 78.5 و78.9% لسرعتي التقليب الميكانيكي 250 و200 لفة/ الدقيقة على الترتيب. أعطى التقليب الميكانيكي عند سرعة 150 دورة في الدقيقة أفضل الظروف لعمل محلول الجير المطفأ الذي ظل مستقرا من حيث بقاء محتوى غاز الميثان فوق نسبة 75٪ لمدة وصلت الي 10 ساعات مقارنة ب 9 ساعات عد سرعات التقليب الميكانيكي 250 و200 لفة/ الدقيقة و5 ساعات عند التقليب اليدوي. لذا، نوصي باستخدام التقليب الميكانيكي بسرعة 150 لفة/ الدقيقة لمحلول الجير المطفأ 50٪ مع هيدروكسيد الصوديوم 10٪ و20٪ على الترتيب لإزالة ثاني أكسيد الكربون وكبريتيد الهيدروجين من الغاز الحيوي حيث انه اعطي اعلي نسبة ميثان واقل نسبة ثاني أكسيد الكربون وحافظ على كبريتيد الهيدروجين عند الحدود المرغوبة والأمنة.