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RESEARCH ARTICLE

CO_2 AND H_2S REMOVAL FROM BIOGAS BY USING FOUR MEMBRAN OF INTEREST

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Abstract

producers of small installations due to the high cost of acquisition of all-in-one installations designed and used in developed countries. This study brings a clear idea on some purification methods accessible to these small producers while proposing those optimal for decarbonation and desulphurization of biogas. Starting from the analysis over time of the biogas resulting from the anaerobic digestion of cow dung, the system of NaOH filters and steel wool mounted in series is the one proposed for optimality (with purification capacities respective CO_2 of the order of 95.9063 ± 0.0864 % and H_2S of 100 %). Better still, the use of a steel wool filter in series with activated carbon (based on coconut shells or bamboo) makes it possible to increase the desulfurization capacity of the latter up to 100 %.

Introduction:-

Biogas upgrading in Africa remains a huge challenge for

The unfortunately exhaustible fossil resources are constantly being exploited to meet the ever-increasing energy needs of populations all over the world (Statistical Review of World Energy, 2022). Biogas, a renewable energy, stands out as a valid alternative to resolve this situation. However, the use of the latter in raw form causes enormous damage in biogas engines and equipment. It therefore appears necessary to rid it of harmful compounds (Tira and Padang, 2016). Many techniques exist and vary according to the compound(s) to be eliminated, the purification capacity, the energy requirement, the cost: these are physical or chemical adsorption methods, biological processes, dehumidification, and global biogas purification technologies.

Physical adsorption includes the use of activated carbons, zeolites, metal oxides… (Chouikhi, 2020) while chemical adsorption involves solutions of amines, sodium hydroxide, calcium hydroxide, hydrogen peroxide etc. (Dubosis and Thomas, 2010). Biological processes include the addition of in-situ air in the biodigesteur (Couturier, 2009), methanogenesis (Hara et *al.*, 2013) and the bio scrubber, among others. Dehumidification methods include physical drying methods (water traps, demisters, cyclone separators) and chemical drying methods (metal oxides, chemical absorption on glycol). These different techniques allow the purification of hydrogen sulphide (H_2S), carbon dioxide (CO_2) or water vapor (H_20) to a significant extent. They can be combined according to the means available but also the need for purification sought in order to obtain a more efficient system.

Global biogas purification technologies or all-in-one technologies are sophisticated devices that are more efficient and useful for the purification of raw biogas because they promote the purification of a wide range of harmful compounds.

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In Benin, producers struggle to obtain them because of their very high cost and the specific conditions they require. For these small biogas producers, the only way out is the adoption of simple techniques for the purification of the biogas produced.

However, we note the virtual absence of studies specific to the use of these methods in Benin. Therefore, having reliable data could help production sites to certify the quality of purified biogas or even allow their purification techniques to be improved. This is the goal of this study, which determines an optimal model for the purification of biogas from materials obtained in the country.

Materials and Methods:-

The experiments are carried out in the premises of BIOGAZ BENIN SARL (Calavi-Benin) from a fixed 8 m^3 biodigester, a purification circuit equipped with filter columns, a gas analyzer and membranes of purification to be tested. Continuous type, the cylindrical-shaped biodigester with a manual stirrer is fed every 2 days with a substrate of cow dung. It therefore allows the production of the biogas necessary for the series of experiments to be carried out. The filters are 110 Ø pipe columns each measuring 0.5 m in length. Hermetically sealed with two 110 Ø plugs to allow the only circulation of the biogas, a column contains a specific membrane allowing the purification of the biogas. The top cap of each column is pierced with O_2 holes on which a 15 Ø 15 nut is placed: one of the holes will be used for the entry of the biogas into the column while the other will allow its exit. To force the biogas to reach the bottom of the column before coming out, a 20 Ø pipe 0.4 m long is connected to the nut of the hole used for the biogas inlet. The analyzer used for biogas composition measurement is a Portable Composite Detector Model: TY-PCD-3, S/N: 21G09040 with gas detection ranges such as CH_4: 0 - 100% VOL, CO_2: 0 - 100% VOL and H_2S: 0 – 1000 PPM designed by SHENZHEN TEENWIN ENWIRONMENT CO., LTD.

The purification membranes chosen for our tests are steel wool, activated carbon based on coconut shell and another based on bamboo, then soda pellets to make a solution of soda hydroxide (NaOH). Four (04) standard filter columns are made for either H_2S or CO_2 purification. Each column is duplicated for the sake of repeatability of experiments and conformity of data (Table 1).

Table 1:- Composition of each type of filter column made.

Column /Indications	NaOH	Steel wool	Type ‘’C’’ activated carbon	Type ‘’C’’ activated carbon
Mass (g)	120	630	40	40
Volume (L)	1	/	/	/

Two (02) purification circuits are to be considered in the context of our study: one comprising a filter column and the other two filter columns in series (Figure 1 and Figure 2). In addition to this point of divergence, each purification unit consists of a raw biogas inlet into the circuit, which is also a point of analysis for this gas coming from the biodigester. Once in the circuit, the biogas can pass through one of the purification lines depending on the filter column(s) to be tested. A gas measurement point at the outlet of the filter column(s) allows analysis of the purified biogas as it travels to the storage tank.

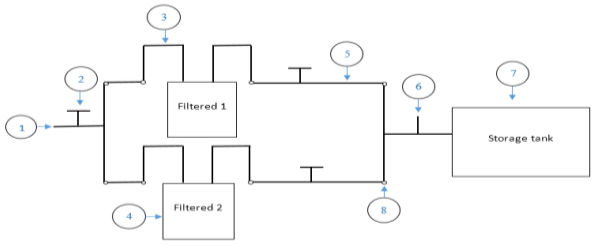

Figure 1:- Diagram of the biogas purification circuit (test from a single column

(1) Biogas inlet from the digester and raw biogas measurement point; (2) Shut-off valve; (3) Washbasin hose; (4) Filter column; (5) 20Ø PVC pipe; (6) Purified biogas measurement point; (7) Purified biogas storage tank; (8) 20Ø elbow (Figure 2 below).

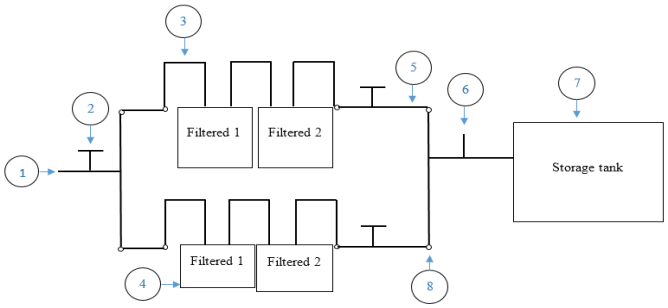

Figure 2:-Diagram of the biogas series purification circuit (test from series columns)

Operating mode:-

At each purification test, the data of the raw components of the biogas such as CO_2, H_2S and CH_4 are taken using the gas analyzer available on site. Once the composition of the biogas at the circuit inlet is known, the main supply valve for the entire purification circuit is opened, then the valve of the purification line to be tested. The biogas then comes into contact with the compound of the filtering column present on the line and undergoes purification. Finally, the line outlet valve is opened for analysis of the biogas components at the outlet at the measurement point made for this purpose.

Measurements:-

Measurements are taken over time and each analysis lasts 60 s. All the results presented in this document come from several experiments in order to see the repeatability of the data; the results presented below are representative of several tests. Once the biogas has been analyzed at the inlet and at the outlet of a purification unit, the purification capacity of the membrane tested for the compound to be eliminated is calculated and expressed in %.

Test condition:-The biogas leaves the digester at a pressure which varies between 1 and 4 KPa and a temperature of 28°C. All the measurements are made at ambient temperature which varies between 30°C and at atmospheric pressure. Presentation of the circuits carried out. The photos below show the completed purification unit (Figure 3 and Figure 4) below.

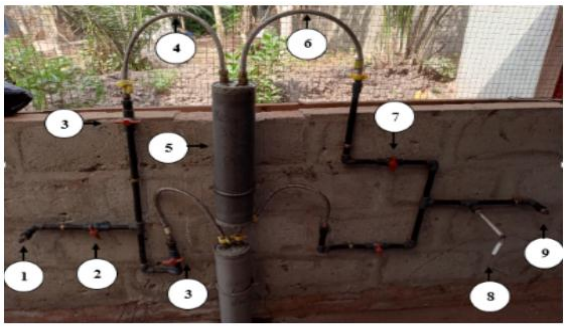

Figure 3:-Purification circuit from a single filter

Figure 4:-Purification circuit from 2 filters in series

1-Biogas inlet from the digester and raw biogas measuring point; 2-Biogas inlet/shut-off valve in the purification unit; 3-Biogas inlet/shut-off valve at the inlet of the purification line; 4-Flexible washbasin (inlet to the purification system); 5-Filter column; 6-Flexible washbasin (outlet into the purification system); 7-Biogas inlet/shut-off valve at the outlet of the purification line; 8-Cleaned biogas measuring point; 9-Purified biogas outlet to the storage tank.

Results and Analysis:-

Decarbonation with a solution of sodium hydroxide (NaOH)

The results of the experiments carried out with the NaOH filter (3 mol/L solution) are shown in Figure 5. It can be seen that the concentration of CO_2 in the raw biogas reaches a maximum threshold of 99.95%Vol. Purification with the NaOH filter leads to a decrease in this concentration down to 4.09%Vol, as reflected in the decreasing trend of the purified CO_2 curve. The calculated average CO_2 removal capacity is 95.91%.

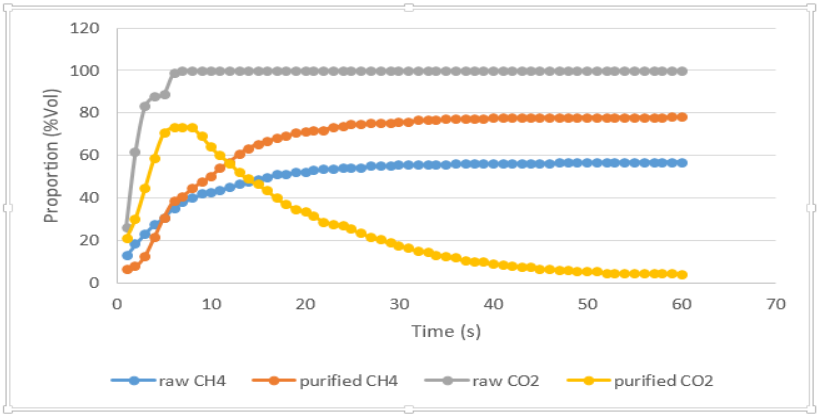

Figure 5:-Average purification of raw biogas with NaOH and observations on CO_2 and CH_4

This value is close to that found (93.16%) following the work of (Tippayawong and Thanompongchart, 2010) with a 0.1 M NaOH solution in a pyrex glass cylinder 70 mm in diameter and 1 m in height. height in a plastic bubble column. The adsorption column was filled randomly with a packing material and the tests were carried out at an ambient temperature of 30°C and 0.1 MPa. We therefore understand the reason for the high purification capacity observed since the packing materials have made it possible to increase the contact surface between the NaOH solution and the biogas for efficient absorption of CO_2. (Zhao et *al.*, 2010) instead report a purification capacity of 85% with a 10% NaOH solution in a polyethylene column 1.8 m high and 300 mm under identical temperature and pressure conditions. One could say that the particular presentations of each purification column as well as the flow rates used could be the basis of the differences observed.

In the same figure, a difference can be seen between the proportion of crude CH_4 and that purified. Indeed, the curve showing the proportion of purified CH_4 experiences an increase compared to that crude from the 6th second. This increase in the present study reaches a maximum of 21.45%Vol.

A return analysis of the purified CO_2 curve shows the point at the 7th second (73.29%Vol) as the upper extremum with a small fluctuation vis-à-vis the 6th second (73.07%Vol). It could be deduced that the proportions of CO_2 and CH_4 in the biogas are linked and the purification of CO_2 in the raw biogas leads to an increase in the proportion of CH_4 in the purified biogas.

Moreover, (Tippayawong and Thanompongchart, 2010) confirm this hypothesis and indicate a 42.4% increase in the proportion of CH_4. Q. Zhao et al note a 34% enrichment in the proportion of CH_4. It is easy to understand that the more the proportion of CO_2 decreases in the biogas, the more that of CH_4 increases.

1. Désulfurisation avec un filtre de laine d'acier, de charbon actif de coque de coco ou de charbon actif de bambou
2. Biogas purification using single purification columns.

In this first section, it is a question of testing the respective purification capacities of steel wool, activated carbon C and B. For the raw biogas analyzed at the inlet of each filter column, there is a regular trend in the proportion of H_2S which reaches a threshold of approximately 220 ppm. Following the experiments, we note that the purification with steel wool brings out the purified biogas with a proportion of H_2S of 0 ppm, i.e. a total purification capacity. Activated carbons C and B purify the biogas to 2.1 ppm and less than 4 ppm respectively. These results show that steel wool is an excellent H_2S purifier compared to activated carbons (Figure 6 and Figure 7 below).

(Antonio-Abdu Sami et *al.*, 2017) found for a flow rate of 3-4 L/min and a mass of 1.9 kg of steel wool, a proportion approaching 0 ppm after passage of the raw biogas in the first filter column (the purification unit being 3-stage) with a starting proportion of 150 ppm from a fixed bed of regenerated steel wool. (Riyadi et *al., 2018)* obtained an average purification capacity of 97% from a 100 cm filter column and a flow rate of 0.1 L/min. The same study demonstrated that the height of the stripping column in the study of the H_2S purification capacity does not have a significant impact on the adsorption of H_2S.

(Kulkarni and Ghanegaonkar, 2019) reinforce this hypothesis even more because their experiments show that variations in flow and pressure have a less significant effect on the efficiency of H_2S removal. The purification capacity found for wool is therefore consistent with these data.

Concerning the purification of H_2S with activated carbon made from coconut shells, (Zulkefli et *al.*, 2019) obtained an adsorption capacity of 0.582 mg H_2S/g. (Orhorhoro et *al.*, 2018) obtained a purification capacity of at least 98%. This validates the purifying capacity found in our study.

The other point to emerge from the graph is that activated carbon B has a lower purification capacity than activated carbon (C). However, the information found in the literature is insufficient to validate or invalidate this hypothesis. Indeed, it should be known that until a very recent past, activated carbons were used only to filter water as well as for fertilization and soil amendment. All of this explains the lack of information observed.

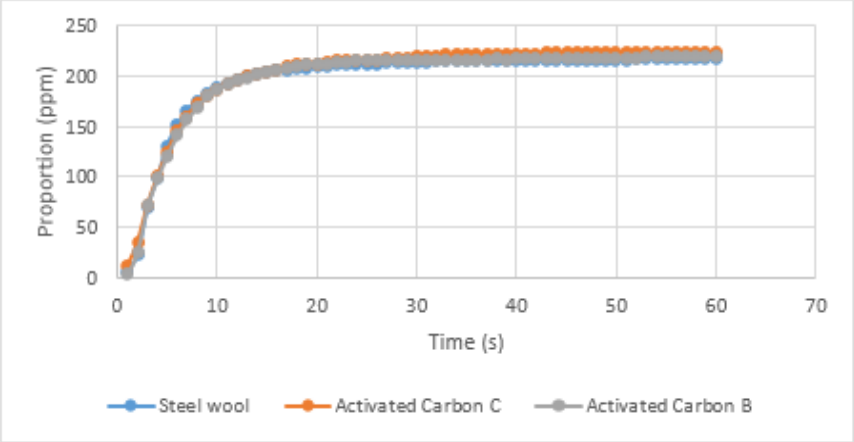

Figure 6:-Proportion of H_2S in the raw biogas at the inlet of each filter column

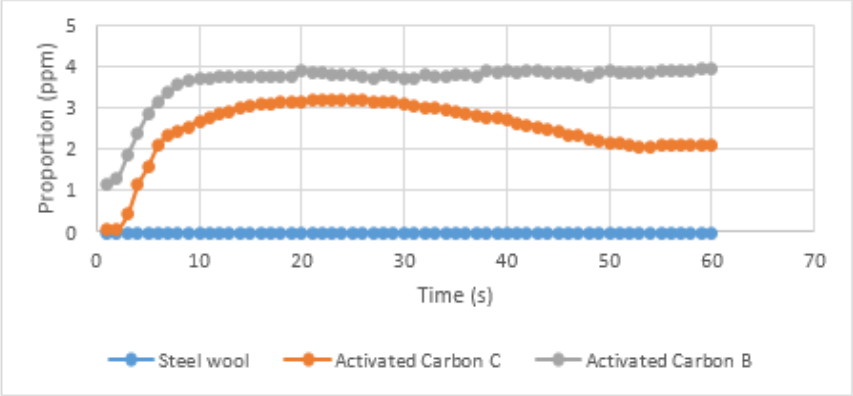

Figure 7:-Proportion of H_2S in the biogas at the outlet of each filter column

Biogas purification using multiple purification columns

Here, two (02) filter columns composed of steel wool and a type of activated carbon are arranged in series for the desulphurization of raw biogas. Importance is given in our experiments to the arrangement order of the filters as well as to the purification capacity of each unit. Also, the use of each type of activated carbon in juxtaposition with a steel wool filter for the desulfurization of biogas shows the originality of this study. From the raw biogas analysis results in this section, we again notice that the proportion of H_2S follows the same trend. Thus, for a maximum starting composition of 212 ppm, the biogas comes out of each purification unit (wool filters & activated carbon in series in a given order) purified to 100%. A total purification observed contrary to that noted during the individual purification tests with activated carbons C and B. It could be noted that purification with filter columns in series allows better purification of harmful compounds in the biogas and more specifically H_2S in this case. Indeed, the biogas purified in the first filter would still undergo purification in the second filter: the purification of impurities is therefore reinforced. The total purification capacity noted for the filters in series would be due to the large purification capacity of the steel wool noted previously. It should be noted that this membrane with high purification performance was able to cover the lack of a second, weaker membrane, regardless of the position it occupied in the purification unit. For example, the high purification capacity of steel wool reinforces that of activated carbon C (which has a purification capacity of 99.09%) so that the purification capacity of the two filters in series is close to the 100% (Figure 8 and Figure 9 below). The experiments reported in the literature do not emphasize the order of the filter columns. However, carrying out the same experiment with a commercial activated carbon led to a purification yield of 96.83% (Riyadi et *al.*, 2018). In the

same study, the desulfurization capacity of each of the two adsorbents was expressed at 92.4%. The hypothesis of greater efficiency of purification filters arranged in series is verified.

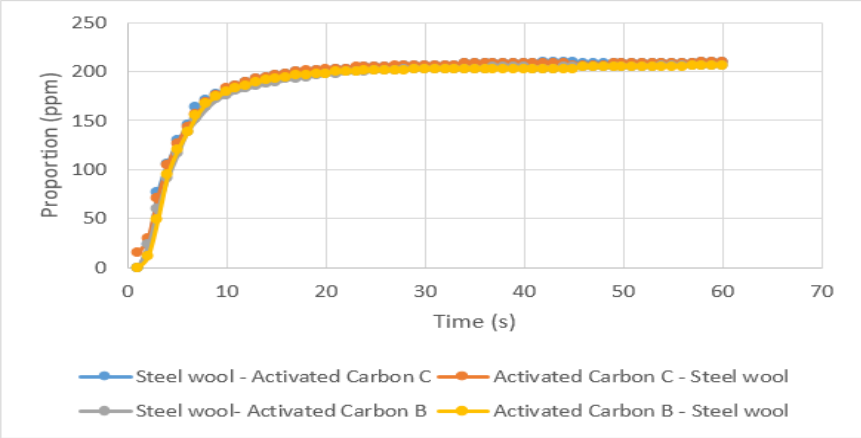

Figure 8:-Proportion of H_2S in the raw biogas at the inlet of each purification unit

A purification unit mounted in series and ordered this time so that the raw biogas passes through filters (3) of wood shavings, steel wool and activated carbon has made it possible to obtain a gas with a proportion of H_2S approaching 0 (Orhorhoro et *al.*, 2018). This result is the same as that obtained for the experiments carried out in this section. We could qualify these purification units as perfect since a total desulfurization has been carried out.

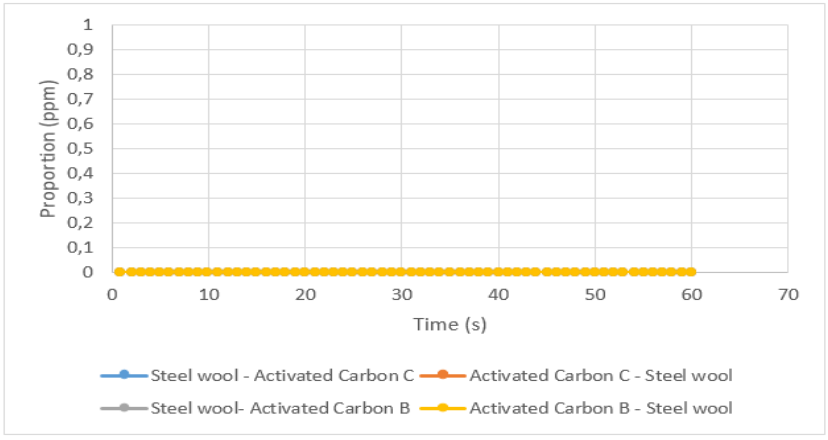

Figure 9:-Proportion of H_2S in the biogas leaving each purification unit

In the works of (Mamun and Torii, 2017) several scrubbing systems are tested for desulfurization with biogas produced from cafeteria waste, vegetables, fruits and livestock manure. The filter system composed of solid lime (CaO), activated carbon and sodium sulphate (Na_2SO_4) allowed a maximum reduction of the proportion of H_2S of 95.9%. This is followed by those of solid lime, zero-valent iron (Fe^{2+}) and sodium sulphate, then solid lime, iron oxide (Fe_2O_3) and silica gel with a respective purification capacity of 90% and 89%. All these purification capacities are lower than those obtained with the activated carbon & steel wool system proposed here. However, the purification capacities obtained are within the same range.

Conclusion:-

The biogas used here comes from the anaerobic digestion of cow dung. Following tests carried out using a solution of NaOH, steel wool and activated carbons (based on coconut husks and bamboo), data concerning the desulfurization or decarbonation capacity of these membranes are available. Thus, we notice that the steel wool and the NaOH solution respectively have a strong desulphurization and decarbonation capacity of the raw biogas. A NaOH filter and a steel wool filter mounted in series therefore become the optimal purification solution proposed.

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