Proposition of biogas filtration system

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Abstract:
The improvement of the calorific value of the biogas, requires purification for removing CO₂ and H₂S. In this study, we present the kinetics of biogas purification by the KOH solution which allows a fast and complete removal of CO₂ and H₂S and by the rust (oxidized iron) which removes only, and quickly H₂S. Consequently, the construction of a filtration system which combines the two substrates (rust and KOH) would increase the calorific value of biogas by eliminating of non-flammable gases (CO₂ and H₂S) and protects cogeneration engines. We propose a conception of the purification system for laboratory tests.

Key words: Biogas, Calorific value, Energy, Filtration system, Purification.

1. Introduction:
The anaerobic digestion consists in the transformation of the organic matter present in the waste into biogas as green energy. Biogas is a mixture of gases (CO₂, CH₄, H₂S, H₂...) [1]. The quality of biogas is an important parameter for his energetic and economic valorization. The biogas is composed from about 55 to 70% of CH₄, 30 to 45% of CO₂, and traces of NH₃ (80 to 100 ppm), H₂O (1000-3000 ppm) and other (100 ppm) [2]. Methane has an energetic interest because it has a high calorific value of the order of 9.94 kWh/m³ [3]. CO₂ is a gas without energetic interest and his presence in the biogas in large quantities can reduce its energy value [4]. Hydrogen sulfide is a corrosive element for motors cogeneration, consequently it risks of destroying them and corrode pipes of station for methanation [5].

A biogaz of good economic and energy quality must contain a high content of methane and a small amount of CO₂ and must be without H₂S. At the industrial scale, the valorization of biogas needs a passage in a purification phase [6]. So, the purification of biogas has become a requirement to obtain a good biogas quality with a high calorific value and without of corrosive impurities.

In this study, we seek to compare solutions and the materials abundant in the Moroccan market, that remove CO₂ and H₂S from biogas produced without affecting its methane content. This will allow us to provide an efficient and less expensive filter for purification of biogas in biogas plants nationwide. So this work is part of the first attempt to study the technical feasibility of this concept and its implementation.

2. Materials and methods:
2.1. Origin and composition of biogas:
The biogas used in this article is harvested from a pilot digester of 1000 liters capacity installed in our university. This digester produces biogas from to the manure of broiler chickens. For each test of purification, we filled an inner tube (KABAT 16.9.28) into biogas and we determined its initial composition by using the portable analyzer (Geotech biogas, Model 5000). This biogas is named raw biogas (before purification).

2.2. Filters:
In this study, we used different supports to evaluate their effectiveness:
- Potassium hydroxide (KOH) with a concentration of 2 N and a pH (13.56 ± 0.1).
- Tap water obtained directly from the laboratory with a pH (7.32 ± 0.1).
- Rust of the steel wool.
- Sawdust of wood.

The pH value is measured using a pH meter of Consort type, calibrated at pH 4 and 7 [7].
2.3. Test of purification:
2.3.1. Experimental device:
The experimental device is constituted of an inner tube filled with raw biogas, it is connected to a valve for flow control (2-lane valve PVDF) then to a digital flow meter (McMillan flow products, Model 50D-7, Air 0-1000 ml / min) for displaying, controlled and verified the flow of raw biogas entering the filtration system (Fig. 1).

The filtration system consists of one bottle (Schott Duran). It is hermetically closed by a silicone plug with two holes (incoming and outgoing of biogas). The first hole is occupied by a pastor pipette to reduce the flow of incoming biogas and to increase the time of contact with the solution or material tested for purification by created of small air bubbles. The second hole of this bottle is connected with the pipe to a valve of four-lane (4-lane valve PVDF):

- The first lane ($V_1$) corresponds to the arrival of the biogas.
- The second lane ($V_2$) is connected to an inner tube (ATK 24x1.95-2.125) for storing the purified biogas.
- The third lane ($V_3$) is connected to the biogas analyzer (Geotech biogas Model 5000).
- The fourth lane ($V_4$) is connected to the outside for emptying.

At the output of the fourth lane, we have installed a valve (valve 2-lanes PVDF) for empty the inner tube of storage. For the rust, we used a bottle of 50 ml. For solutions (KOH, water and sawdust), we used a larger bottle (250 ml) for improving the reaction time.

![Figure 1: Experimental device of purification test.](image)


2.3.2. Experimental method:
After determining the initial composition of raw biogas, we open the valve for flow control so that it appears constant flow on the flow meter ($120 \pm 5$ ml / min). Then the raw biogas passes into the bottle and it is bubbled through the purification system during 20 minutes, it becomes then a biogas purified that we store in an inner tube of storage (Fig. 2).

After each 20 minutes of bubbled, we close the valve flow and we open the valve of four lanes toward biogas analyzer which records the final composition, then we empty the inner tube of the storage for removing the residual biogas. Finally, we did the same process every 20 minutes for two hours.

2.4. The calculation of lower heating value and yield of purification:
The methane is a flammable gas with a lower heating value (LHV) in the order of 9.94 kWh / m³. Therefore, the LHV of biogas is proportional to its methane content which can be calculated according to the following equation [3]:

\[
LHV = \frac{9.94 \times \text{methane content}}{\text{m}^3} \]
LHV (Kwh/m³) = Percentage of methane in biogas / 100 x 9.94
For characterizing a substrate for biogas purification, it must determine the yield of purification.

Figure 2: Sense of displacement of biogas in the device.

The effectiveness of different substrates in the capture of CO₂ and H₂S, is not the only performance criteria, because the filter should not affect the composition of CH₄, so the best result would be to eliminate the non-flammable gases (CO₂) and the corrosive gas (H₂S) without any influence on the content of the methane. This characteristic is determined by the following equation:

The yield of purification (%) = \[\frac{\text{LHV}_{\text{purified biogas}} - \text{LHV}_{\text{raw biogas}}}{\text{LHV}_{\text{raw biogas}}} \times 100\]

3. Results and discussion:

3.1. Composition of raw biogas:
In our digester, we obtain a raw biogas mainly composed of methane (72.7 ± 2.9%) followed by CO₂ (27.1 ± 2.7%) and hydrogen sulfide (42.5 ± 10.6 ppm) (Tab.1). This biogas has a LHV of 7.2 kWh / m³. The origin of this biogas is the anaerobic digestion of droppings of broilers.

Table 1: The composition of raw biogas.

<table>
<thead>
<tr>
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<th>Raw biogas</th>
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<tbody>
<tr>
<td>CH₄ (%)</td>
<td>74.8 – 70.6</td>
</tr>
<tr>
<td>CO₂ (%)</td>
<td>29.1 – 25.2</td>
</tr>
<tr>
<td>H₂S (ppm)</td>
<td>50 – 35</td>
</tr>
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</table>

3.2. Kinetics of biogas purification:
After two hours of testing, we note that the effectiveness of the purification of biogas is different depending on the substrate (Fig. 3 to Fig 6):

- The methane (CH₄):
  Only two substrates, KOH and rust, don’t affect the methane content in biogas during the purification process, but other substrates, water and sawdust, decreased its content in the biogas. Therefore, these substrates have decreased the calorific value of the purified biogas.
- The carbon dioxide (CO₂):
KOH solution is the only substrate which allows a fast and complete removal of CO₂, but the others substrates used are unable to eliminate this gas. This result is due to the capture of CO₂ in the KOH solution according to the following chemical reaction [8]:

\[
\text{KOH} + \text{CO}_2 \rightarrow \text{KHCO}_3 \\
\text{KHCO}_3 + \text{KOH} \rightarrow \text{K}_2\text{CO}_3 + \text{H}_2\text{O}
\]

Some researchers have used this purification by KOH to produce biogas measuring devices [9,10]. Therefore the removal of CO₂ by KOH with a concentration of 2N is a better technique for purifying biogas compared to other studies which proposes an elimination with a 40% KOH solution in three steps [11].

- The hydrogen sulfide (H₂S):
The rust is the only substrate that removes only and quickly the H₂S. The adsorption of H₂S at iron oxide can be illustrated by the following equation [12]:

\[
\text{Fe}_2\text{O}_3 + 3\text{H}_2\text{S} \rightarrow \text{Fe}_2\text{S}_3 + 3\text{H}_2\text{O}
\]

The water can absorbed the H₂S according to reactions [13]:

\[
\text{H}_2\text{S} + \text{H}_2\text{O} \rightarrow \text{H}^+ + \text{HS}^- \\
\text{HS}^- \rightarrow \text{H}^+ + \text{S}^{2-}
\]

But the purification of biogas by the H₂O is undesirable because it can generate oxygen. However, the water is recycled for each purification, the cost increases [14]. The H₂S has an unpleasant odour; at higher concentrations, it can be life-threatening. The recommended industrial exposure limits are from 8 to 10 ppm for 8 hours a day per week [15]. Some authors proposed the iron-chelated solution in water for exemple the chemical absorption in an iron-chelated solution catalyzed by Fe/EDTA [16].

**Table 2: Comparison of biogas purification by different substrates.**

<table>
<thead>
<tr>
<th></th>
<th>LHV of raw biogas (kWh/m³)</th>
<th>LHV of purified biogas (kWh/m³)</th>
<th>Yield of purification (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Purification by rust</td>
<td>7.2 ± 0.2</td>
<td>7.2 ± 0.2</td>
<td>0</td>
</tr>
<tr>
<td>Purification by KOH</td>
<td>7.1 ± 0.2</td>
<td>9.8 ± 0.2</td>
<td>+ 38</td>
</tr>
<tr>
<td>Purification by sawdust</td>
<td>7.4 ± 0.1</td>
<td>6.8 ± 0.1</td>
<td>- 8.1</td>
</tr>
<tr>
<td>Purification by water</td>
<td>7 ± 0.1</td>
<td>6.8 ± 0.1</td>
<td>- 2.8</td>
</tr>
</tbody>
</table>

Consequently, the purification of biogas by KOH solution is the most practical for remove CO₂ and H₂S follow of the rust that quickly removes H₂S and retains the other gases of biogas [17]. The construction of a filtration system, which combines the two substrates, rust and KOH, allows to increase the calorific value and produce a biogas without corrosive element [17,18,19]. In this paper, we present a conception of a filtration system model, it remains to evaluate its performance a function of time and to determine the saturation time for possible transposition in industrial scale.
3.3. The conception of a biogas filtration system:

In this paper, we propose a biogas filtration system which consists of two filters connected by PVC pipe (Figures 7 to 10):
Figure 7: Industrial model of the first filter. Figure 8: Industrial model of the second filter.

The first filter is known as sieve filter. It consists of several sieves in stainless superposed on each other. Inside the each sieve, we have put the rust. When the biogas produced by digester, enters the top of the filter, it is dispersed in the rust which is in the superimposed sieves. This filter quickly eliminates H₂S thanks to the absorbing power of rust for to avoid saturation of KOH by this gas [12]. Consequently, we obtain a biogas without corrosive elements [15]. Thereafter the biogas enters in the second filter for the final purification [17].

Figure 9: The lateral view of the filtration system. Figure 10: The section view of the filtration system.


The second filter is called a bubbler in KOH [17]. It consists of a PVC column occupied below by a conical base with holes. The column and base are immersed in the solution KOH. The biogas produced by the first filter enters the column and spell in the conical base producing the bubbles dissolved in the KOH solution. This filter quickly and completely removes the CO₂ [8,9]. It produces a biogas with high methane concentration and
without of CO₂ and H₂S. Finally, this biogas produced is moving towards a cogeneration engine or to a burner to produce electricity or thermal energy [17].

Conclusion:

From this study, we can conclude that the purification of biogas by KOH and rust allows:
- A quick removal of H₂S and CO₂ without affecting the methane content in the biogas.
- An increase of LHV in the biogas produced.
- A total and rapid elimination of corrosive element.

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