Protection of the Purification Station Thermal Engines Against the Effects of Hydrogen Sulfide by the Coupling of a Chemical and Biological Treatment of the Produced Biogas

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ABSTRACT
Among the various techniques used to reduce hydrogen sulfide in biogas avoiding harmful effects on engines, the chemical and biological treatment appears particularly promising. The main objective of this article was to develop a new process to reduce the harmful effect of hydrogen sulfide (H2S), contained in the biogas resulting from methanization, on the equipment of the wastewater treatment plant (WWTP) of FES city in particular the two cogeneration units. A multiple regime technique for biogas desulfurization, based on the chemical and biological treatment as well as internal micro-aeration of the digester was developed. Owing to the insights gained from this study, it was identified that reducing the concentration of H2S in biogas and improving methane production (biogas production increased from 3.6 M Nm3 in 2018 to 3.8 M Nm3 in 2019, a saving of about 300,000 MAD); reduction of desulfurization tower downtime from 4 times/year to 1 time/year; increasing operating time of generating sets from 8800 in 2018 to 14 400 h in 2019; electricity production increased from 5.9 GWh in 2018 to 7.2 GWh in 2019. In light of these findings, it can be affirmed that the study successfully achieved its objectives, presenting valuable avenues for future scientific exploration

Keywords: water treatment plant, anaerobic digestion (AD), biogas treatment, biological desulphurization, micro-aeration and thermal engines.

INTRODUCTION
Today, renewable energies represent a primary interest in the face of alarming energy consumption and environmental risks. Among the production techniques of these renewable energies, one can cite anaerobic digestion (AD), or methanization, which has many environmental, economic and social interests. Anaerobic digestion is a microbiological technique for converting organic matter into biogas in an oxygen-depleted environment, mainly in the presence of anaerobic microorganisms. This biogas can be recovered in the form of heat or electricity (Szczyrba et al. 2020). The positive environmental impacts of anaerobic digestion, such as the reduction of greenhouse gas emissions and odors, are widely used as strengths for the promotion of this process. Indeed, the biogas from AD is a mixture of methane (50–70%), CO2 (30–50%), H2 (1–5%), H2S (0.1–3%), N2 (2–7%) and sometimes NH3 (Kapoor et al. 2019). These percentages depend essentially on the quality of the substrate and on several parameters of the operation digester (Id 2015). The upgrading of biogas CH4 biogas in the production of electricity and heat remains limited due to the presence of pollutants, especially H2S, which has harmful effects on equipment and heat engines (Okoro et al.2019). Anaerobic digestion or methanization is a microbial fermentation based on the degradation of organic matter in a reactor called a digester, in absence of oxygen and under fairly specific conditions (Quality of the substrate, T °, pH, length of stay, etc.) (Hajaji et al. 2016). This microbial
degradation releases a biogas composed mainly of methane (CH\textsubscript{4}) and carbon dioxide CO\textsubscript{2} (Figure 1). It also contains compounds, such as hydrogen sulfide (H\textsubscript{2}S), ammoniac (NH\textsubscript{3}) and other volatile organic compounds at low concentrations (Kapoor et al. 2019).

This reaction process is classically divided into four biochemical steps (Ghouali et al. 2015, Meres et al. 2009):

− hydrolysis: organic macromolecules (polysaccharides, proteins, lipid compounds) are hydrolyzed into simpler elements, such as simple sugars, amino acids, short-chain fatty acids, glycerol;
− acidogenesis: the substrates resulting from the hydrolysis phase are transformed by bacteria into volatile fatty acids, alcohols, ammoniac, carbon dioxide and hydrogen;
− acetogenesis: volatile fatty acids (other than acetic acid) are converted by acetogenic reducing bacteria into acetate, hydrogen and carbon dioxide;
− methanogenesis: the substrates resulting from the hydrolysis phase are reduced by bacteria to methane and carbon dioxide.

At the end of these successive reactions, biogas (composed of CH\textsubscript{4}, CO\textsubscript{2}, H\textsubscript{2}S, etc.) as well as a stabilized and hygienized digestion residue, called digestate (sludge), are formed. H\textsubscript{2}S is the result of the fermentation of sulfur compounds present in many organic materials (Aziza et al. 2012. It is produced by two types of sulfur-reducing bacteria (SRB). The latter use either acetic acid or propionic acid as organic substrate to reduce sulfates (SO\textsubscript{4}\textsuperscript{2-}) to sulfides (H\textsubscript{2}S and HS\textsubscript{-}). The chemical reactions involved are represented by Equations 1 and 2, (Boivin et al. 2010):

\[
\begin{align*}
H_2SO_4 + CH_3COOH & \rightarrow H_2S + 2 H_2CO_3 \quad (1) \\
H_2S_4 + 4/7CH_3COOH & \rightarrow H_2S + 12/7 H_2CO_3 \quad (2)
\end{align*}
\]

From the point of view of the digester feed, the oxidized forms of sulfur are found in:

- minerals in the form of sulfates (for example: in many vinasses resulting from the regeneration of ion exchangers with sulfuric acid);
- organic matter: like proteins.

H\textsubscript{2}S causes several problems when it is present in the biogas recovery circuit resulting from anaerobic digestion (Ramos et al. 2014). As expressed by equations 3 and 4, the combustion of H\textsubscript{2}S produces sulfur dioxide (SO\textsubscript{2}) which itself, when oxidized, produces sulfur trioxide (SO\textsubscript{3}). The latter compound reacts chemically with water to form sulfuric acid, a strong acid which gives a corrosive potential to combustion biogas (Eq. 5) (Boivin et al. 2010):

\[
\begin{align*}
H_2S + 3/2 O_2 & \rightarrow SO_2 + H_2O + 518 \text{ kJ/mole} \quad (3) \\
SO_2 + 1/2 O_2 & \rightarrow SO_3 + 99 \text{ kJ/mole} \quad (4) \\
SO_3 + H_2S & \rightarrow H_2SO_4 \text{ (gas)} + 101 \text{ kJ/mole} \quad (5)
\end{align*}
\]

The presence of H\textsubscript{2}S and water vapor in the biogas causes premature wear of combustion equipment and also alters the metal structures that surround the anaerobic digestion system. In the case of an internal combustion engine (Cogeneration), the

![Figure 1. Flow diagram of the AD process](image-url)
H₂S reacting with the water will attack the pistons and cylinders. In fact, the H₂S concentration should not exceed 500 ppm to limit damage to internal parts of the equipment (Zhao et al. 2010). Table 1 summarizes the consequences of H₂S contained in the biogas produced in the wastewater treatment plant.

In this context, and faced with all these effects of the H₂S component mentioned previously, and for a better valorization of CH₄ biogas, the authors worked on an important study for the treatment of H₂S, based on the combination between the biological method, the chemical method and a slight micro-aeration of the digester within the wastewater treatment plant in the city of Fez. The main objective of the study was to improve the quality and production rate of biogas, reduce the corrosive impact of H₂S on the equipment of the wastewater treatment plant, and optimize the operating life of the cogenerators and therefore the increase in the clean quantity of electrical energy produced.

MATERIAL AND METHODS

Biological treatment of H₂S in the WWTP-FES

For the treatment and valorization of biogas resulting from anaerobic digestion, within Fez city plant wastewater treatment (Morocco), the biological method (also called: Desulfurization of biogas) is used, which is carried out by biological washing on a biological purifier (Figure 2). This solution makes it possible to lower the H₂S content without producing polluting by-products, as a result, the maintenance frequency of the desulfurization tower will be considerably reduced, which will contribute to improving the efficiency of the station.

This desulfurization of the biogas is done by biological washing in a counter-current contact tower. The biogas flow is ascending, the wash water flow (contains nutrients for the bacteria) to a descending flow. This solution reduces the H₂S content without producing polluting by-products. The biological elimination of H₂S is carried out in the presence of microorganisms and traces of oxygen (at < 25% of the allowed limit concentration) to oxidize the H₂S into sulfate. The final purge, which has a sulfate concentration of approximately 5 to 8% and a pH of around 1.5, will be pumped to the upstream side of the WWTP pretreatment. To maintain sufficient microbial activity, the temperature of the unit is maintained at 25°C using hot water from the cogeneration plant. Basic data of the desulphurization tower:

Table 1. The different harmful effects of H₂S (Zhao et al. 2010)

<table>
<thead>
<tr>
<th>Effects of H₂S</th>
<th>Consequences</th>
</tr>
</thead>
</table>
| **On human health** | * 0.02-0.13 ppm H₂S: olfactory perception.  
* 250-500 ppm H₂S: headache, cyanosis, pulmonary edema.  
* >1000 ppm H₂S: Apnea, nervous system paralysis and death within minutes |
| **On CH₄ biogas production** | The presence of oxidized sulfur (SO₄²⁻) consumes acetic acid and hydrogen and reduces the production of CH₄, according to the following reaction:  
CH₃-COOH + SO₄²⁻ → 2CO₂ + H₂S |
| **On cogeneration engines during combustion** | * Sulphates (HSO₄⁻) form sulfuric acid which attacks aluminum alloy pistons and eventually pierces them.  
* The sulfur precipitates on the valves, decreases the compression and can eventually break the valve heads. |

Figure 2. Synoptic diagram of the desulfurization tower (SCADA-STEP FES software)
• Raw biogas throughput to be treated: 1,440 Nm³/hour
• \text{H}_2\text{S} \text{ in content: Varies between 3,000 ppm and 7,000 ppm.}
• \text{H}_2\text{S} \text{ out content: < 500 ppm}
• Average removal capacity is: 6.5 kg H₂S/h.

However, it was necessary to ensure rigorous monitoring of certain parameters in order to guarantee the reliability of the study for improving the operation of the desulfurization tower and the micro-aeration of the digesters (Awe et al. 2017). Table 2 shows the monitoring schedule for the necessary parameters of the digester and the desulfurization tower operations.

**Problem encountered at the desulfurization tower**

The desulphurization tower must reduce the concentration of H₂S from 4,500 ppmv (average value, in 2018) to less than 500 ppmv (recommended value) without producing dangerous by-products but, the major disadvantage of this method is the appearance of sulfur (S) according to reaction (Equation 6), which is a solid compound and causes a rapid clogging of the tower (Figure 3).

\[
\text{H}_2\text{S} + \text{O}_2 \rightarrow 2\text{S} + 2\text{H}_2\text{O} \quad (6)
\]

The cumulative effects of the clogging of the tower cause an internal increase in the pressure drops between the inlet and the outlet of the biogas, and significantly reduces the efficiency of H₂S removal. This makes the biogas more corrosive to the metal parts of the installation. Regulators, gas meters, valves and supports can quickly corrode. The combustion of biogas containing H₂S produces sulfur dioxide (SO₂). When SO₂ combines with water vapour, it produces sulfuric acid which corrodes the exhaust pipes of engines, etc. Gaseous SO₂ also dissolves in engine oil, causing the oil to become acidic and lose its ability to lubricate, damaging the engine and shortening the time between oil changes (Shab et al. 2013).

<table>
<thead>
<tr>
<th>Table 2. Planning for the required parameters of the digester and the desulfurization tower operations</th>
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</thead>
<tbody>
<tr>
<td>Parameters</td>
</tr>
<tr>
<td>---</td>
</tr>
<tr>
<td>Biogaz flow</td>
</tr>
<tr>
<td>(pH)</td>
</tr>
<tr>
<td>(temperature)</td>
</tr>
<tr>
<td>Chemical oxygen demand COD (\text{COD}_{\text{total}}) (Sludge)</td>
</tr>
<tr>
<td>COD (\text{filtrée/after centrifugation} ) (Sludge)</td>
</tr>
<tr>
<td>Volatile fatty acids VFA (Sludge)</td>
</tr>
<tr>
<td>Suspended matter (SM) / volatile Suspended matter (VSM) (sludge)</td>
</tr>
<tr>
<td>Total nitrogen TN + (\text{NH}_4^+)-N (sludge)</td>
</tr>
<tr>
<td>total phosphorus + (\text{PO}_4^{3-})-P (sludge)</td>
</tr>
<tr>
<td>(\text{CH}_4) et (\text{H}_2\text{S})</td>
</tr>
<tr>
<td>Dwell time (sludge)</td>
</tr>
</tbody>
</table>
Corrective actions:
1. QSR (Quick Sludge Removal) operations that require between 4 and 8 hours of work: removes the sulfur deposit from the desulfurization tower by filling it with water and injecting compressed air.
2. Complete washing of the tower, which takes between 4 and 7 days.

For the year 2018, the periodicity of carrying out these two operations according to the value of ΔP, which must always be less than 5 mbar, is shown in Figure 4. After the analysis of Figure 4 it was noticed that the number of QSR operations and complete scrubbing of the desulfurization tower is still quite high, generating additional operating costs. In 2018, the following were carried out:

- Three (03) QSR operations.
- Two (02) complete washing operations of the tower.

Moreover, these operations cause a total stop of the desulfurization tower, which directly influences the purification of the biogas produced and consequently:
- A stoppage of both cogeneration units due to the unpurified biogas: Hydrogen sulfide H₂S in combination with water vapor in the raw biogas form sulfuric acid (H₂SO₄), which is highly corrosive to the cogeneration engine components.
- Very low self-production of electricity (cogeneration of stored biogas).
- Increase in invoiced electricity consumption (Table 3).

**Figure 4.** Periods of QSR operations and washing of the desulfurization tower

**Table 3.** Monthly electricity consumption of the WWTP for the year 2018

<table>
<thead>
<tr>
<th>Month</th>
<th>Flow rate of biogas produced (m³)</th>
<th>Quantity of electricity invoiced (KWh)</th>
<th>Quantity of electricity cogenerated (KWh)</th>
<th>Cogeneration efficiency (%)</th>
<th>Total energy consumption of the WWTP (KWh)</th>
</tr>
</thead>
<tbody>
<tr>
<td>January-18</td>
<td>29 836</td>
<td>330 225</td>
<td>0</td>
<td>0</td>
<td>330 225</td>
</tr>
<tr>
<td>February-18</td>
<td>81 559</td>
<td>352 276</td>
<td>0</td>
<td>0</td>
<td>352 276</td>
</tr>
<tr>
<td>March-18</td>
<td>99 292</td>
<td>414 372</td>
<td>51 416</td>
<td>11</td>
<td>465 788</td>
</tr>
<tr>
<td>April-18</td>
<td>119 203</td>
<td>295 769</td>
<td>388 931</td>
<td>57</td>
<td>684 700</td>
</tr>
<tr>
<td>May-18</td>
<td>206 086</td>
<td>184 982</td>
<td>565 784</td>
<td>75</td>
<td>750 766</td>
</tr>
<tr>
<td>June-18</td>
<td>290 618</td>
<td>504 902</td>
<td>314 723</td>
<td>38</td>
<td>819 625</td>
</tr>
<tr>
<td>July-18</td>
<td>562 105</td>
<td>607 215</td>
<td>923 541</td>
<td>60</td>
<td>1 530 756</td>
</tr>
<tr>
<td>August-18</td>
<td>550 102</td>
<td>221 520</td>
<td>1 033 751</td>
<td>82</td>
<td>1 255 271</td>
</tr>
<tr>
<td>September-18</td>
<td>534 672</td>
<td>699 160</td>
<td>527 659</td>
<td>43</td>
<td>1 226 819</td>
</tr>
<tr>
<td>October-18</td>
<td>471 860</td>
<td>302 520</td>
<td>886 233</td>
<td>75</td>
<td>1 188 753</td>
</tr>
<tr>
<td>November-18</td>
<td>365 849</td>
<td>263 760</td>
<td>760 222</td>
<td>74</td>
<td>1 023 982</td>
</tr>
<tr>
<td>December-18</td>
<td>282 287</td>
<td>499 472</td>
<td>412 393</td>
<td>45</td>
<td>911 865</td>
</tr>
<tr>
<td>Total</td>
<td>3 593 471</td>
<td>4 676 173</td>
<td>5 864 653</td>
<td></td>
<td>10 540 826</td>
</tr>
</tbody>
</table>
After analyzing the results, it was found that:

- Before each QSR (Figure 5), the self-production of electrical energy decreases although the electrical consumption billed increases, this is because of the reduction in the hourly rate of the operation of the two cogeneration units due to the reduction in the volume of biogas purified (shutdown of the desulfurization tower due to the increase in ΔP).
- After each QSR or washing operation, the desulfurization tower resumes his normal operation and the biogas can be purified and cogenerated.

The biological treatment of biogas from anaerobic digestion by a desulfurization tower remains a beneficial solution from an economic and environmental point of view, owing to the limited use of energy and chemicals. However, the major disadvantage of this process is the internal clogging of the tower due to the deposition of sulfur (Eq. 6) and consequently an increase in the internal pressure at this tower, which has a negative influence on the purification efficiency of the biogas sent to cogeneration units for the production of electrical energy. A biogas that is not sufficiently purified can contain traces of H₂S which causes the degradation of the thermal motors of cogeneration units by two mechanisms: sulfurization where the sulfur attacks directly the metal components of the engine and corrosion where sulfuric acid formed with condensation water erodes metal surfaces (Maizonnasse et al. 2013).

Faced with this repetitive situation and following the technical and financial requirements, a technical design study for the desulphurization unit was established by combining the biological treatment of H₂S with a chemical treatment (chemical oxidation + a slight microaeration of the digesters) by injection external from oxygen to biogas upstream of the tower (Maizonnasse et al. 2013).

**Chemical oxidation study**

The process for injecting oxygen upstream of the desulfurization tower is simple to set up and requires a small initial investment. It consists of the installation of an air injection pump coupled to a probe for measuring oxygen and biogas flow. The automatic regulation of oxygen flow prevents the creation of an explosive mixture (Diaz et al. 2015). Thereby, this process ensures at the same time, a microaeration of the digesters which will improve the production of CH₄ and reduce the appearance of H₂S (Polano et al. 2009).

**Adopted solution**

The proposed solution based on the injection of regulated air upstream of the desulfurization tower (Figure 6) allowed:

a) chemically oxidizing the H₂S to S sulfur. This solid element will be deposited on the biogas pipe upstream of the biological H₂S desulphurization tower (Figure 7), avoiding its frequent clogging and consequently the number of annual shutdowns of this tower will be decreased.

b) creating microaeration digesters to improve the quality of biogas and reduce the production of H₂S is based on the biochemical oxidation of the sulfide to elemental sulfur (S0) or/and sulfate (SO₄²⁻) (Dannesboe et al. 2019).

**RESULTS AND DISCUSSION**

After carrying out several oxygen injection tests upstream of the desulfurization tower, it was
found that the optimal volume of oxygen injected is 1/20 of the volume of biogas produced, this allowed considerably improving the biological desulfurization of biogas within the Fez wastewater treatment plant (Figure 8). It enabled to chemically oxidize H₂S to sulfur (S), ensure microaeration of the digesters and significantly reduce the concentration of H₂S in the biogas mixture. On the one hand, according to the reaction: \( \text{H}_2\text{S} + \text{O}_2 \rightarrow 2\text{S} + 2\text{H}_2\text{O} \), microorganisms oxidize hydrogen sulfide with oxygen molecules and convert the rest of the elements into elemental sulfur and water, this solid compound (sulfur) will be deposited on the pipe before entering the biological desulfurization tower. This deposit allowed significantly reducing the rate of clogging and the variation of the internal pressure of the tower (\( \Delta P < 7 \text{ mbar} \)), and consequently reducing the number as well as the time of the annual operations of the QSR and the washing (Figure 9). On the other hand, microaeration (Jenicek et al. 2010) allowed heterotrophic bacteria to inhibit SRB and minimize the appearance of hydrogen sulfide in the gas mixture inside the digester. The Figure 10 clearly shows this result and explains the reduction in the concentration of H₂S after the injection of air into the digester. (Jenicek et al. 2017).

The analysis of Figure 10 shows that the presence of oxygen in the digesters (by microaeration) oxidizes the sulfide present in the mixture, into
elemental sulfur which will be precipitated and removed with the sludge, which means the fall of the concentration of \( \text{H}_2\text{S} \) after the technique of microaeration adopted in the study to improve the operation of the desulfurization tower. It is also noted that the microaeration improves the degradability of the COD and volatile suspended elements and enriches the quality of the biogas produced (Botheju et al. 2010). According to Table 4 and Figure 11, it can be said that the annual monitoring of the energy consumption of the WWTP, after the study of the new design of the desulfurization tower, showed a very satisfactory energy balance, because the availability of biogas streamlined allowed:

Table 4. Monthly monitoring of the electricity consumption WWTP electricity consumption for the year 2019

<table>
<thead>
<tr>
<th>Month</th>
<th>Flow rate of biogas produced (m³)</th>
<th>Quantity of electricity invoiced (KWh)</th>
<th>Quantity of electricity cogenerated (KWh)</th>
<th>Cogeneration efficiency (%)</th>
<th>Total energy consumption of the WWTP (KWh)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jan-19</td>
<td>274 053</td>
<td>111 080</td>
<td>428 180</td>
<td>79</td>
<td>539 260</td>
</tr>
<tr>
<td>Feb-19</td>
<td>275 503</td>
<td>120 000</td>
<td>565 676</td>
<td>82</td>
<td>685 676</td>
</tr>
<tr>
<td>Mar-19</td>
<td>432 987</td>
<td>136 440</td>
<td>958 795</td>
<td>88</td>
<td>1 095 235</td>
</tr>
<tr>
<td>April-19</td>
<td>391 868</td>
<td>253 725</td>
<td>826 601</td>
<td>77</td>
<td>1 080 326</td>
</tr>
<tr>
<td>May-19</td>
<td>299 758</td>
<td>712 840</td>
<td>526 978</td>
<td>43</td>
<td>1 239 818</td>
</tr>
<tr>
<td>June-19</td>
<td>268 015</td>
<td>673 701</td>
<td>687 293</td>
<td>50</td>
<td>1 360 994</td>
</tr>
<tr>
<td>July-19</td>
<td>423 988</td>
<td>563 982</td>
<td>498 957</td>
<td>47</td>
<td>1 062 939</td>
</tr>
<tr>
<td>August-19</td>
<td>387 538</td>
<td>264 040</td>
<td>653 413</td>
<td>71</td>
<td>917 453</td>
</tr>
<tr>
<td>Sept-19</td>
<td>482 823</td>
<td>330 280</td>
<td>887 254</td>
<td>73</td>
<td>1 217 534</td>
</tr>
<tr>
<td>Oct-19</td>
<td>477 125</td>
<td>388 320</td>
<td>907 916</td>
<td>70</td>
<td>1 296 236</td>
</tr>
<tr>
<td>Nov-19</td>
<td>404 325</td>
<td>187 360</td>
<td>296 781</td>
<td>61</td>
<td>484 141</td>
</tr>
<tr>
<td>Dec-19</td>
<td>389 541</td>
<td>166 480</td>
<td>0</td>
<td>0</td>
<td>166 480</td>
</tr>
<tr>
<td>Total</td>
<td>4 507 524</td>
<td>3 908 248</td>
<td>7 237 844</td>
<td></td>
<td>11 146 092</td>
</tr>
</tbody>
</table>
The recovery of a large quantity of CH4 biogas; the cogeneration of a large amount of electrical energy; minimization of the amount of electricity purchased; The reduction in the rate and frequency of maintenance of biogas circuit equipment and especially the heat engines of cogeneration units. This has a positive influence on the number of hours of operation of these two cogeneration units, because it was possible to go from 8,763 hours of operation in 2018 to 14,418 hours of operation in 2019 (Fig. 12).

CONCLUSIONS

The combination of hydrogen sulfide oxidation upstream of the desulfurization tower with digester micro-aeration showed the following results: protection of cogeneration units and heat engines against the harmful effects of H2S; reduction of the clogging rate of the desulfurization tower; improvement of methane CH4 production; increase of electricity production; increase of equipment service life.

Moreover, the study of the oxidation of hydrogen sulfide upstream of the desulfurization tower with microaeration of the digesters has enabled to obtain promising results, especially in terms of: flow of biogas produced of 4,507.524 m3 in 2019 instead of 3,593.471 in 2018; quantity of electricity invoiced of 3,908.248 kWh in 2019 instead of 4,676.173 kWh in 2018; quantity of cogenerated electricity of 7,237.844 kWh in 2019 instead of 5,864.653 kWh in 2018; annual gain can reach an amount of 300,000 MAD.

It was also noted that anaerobic digestion assisted by partial aeration can serve as a beneficial treatment strategy for the simultaneous treatment of waste and energy production.

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REFERENCES


