Enhancement of Fecal Sludge Conversion Into Biogas Using Iron Powder During Anaerobic Digestion Process

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Abstract: Anaerobic digestion is often used to stabilized and convert organic wastes into methane and biological fertilizer. However, when applied to fecal sludge, it doesn’t yield good methane due to its high content of nitrogen. Here we have conducted anaerobic digestion of fecal sludge in the presence of iron powder (Fe) as electron donor. Results showed that 4822.7 mL CH₄ kg⁻¹ was successfully recovered from fecal sludge in the control. The use of Fe in the anaerobic bio-digester remarkably improved methane yield. Indeed, up to 9933.3 mL CH₄ kg⁻¹ wet sludge was recovered when Fe is properly used (1 g Fe for 400 g wet weight), compared to 4822.7 mL kg⁻¹ in the control. The concentration of methane in the produced biogas increased from 58.0% in the control to 72.5% and 77.6% in the presence of iron powder, respectively at the dose rate of 0.5 g Fe and 1 g Fe per 400 g wet sludge. COD removal efficiency was also greatly improved. 65.5% of COD was removed when excreta was properly spiked with Fe (1g Fe) against 42.2% in the control. This corresponds to an increasing rate of 23%. Furthermore, the presence of Fe in the digesters considerably reduced the odor by trapping produced sulphur ion and prevent the formation of H₂S responsible for the sickening odor.

Keywords: Fecal Sludge, Anaerobic Digestion, Valorization, Iron Powder, Methane

1. Introduction

Fecal sludge known as excreta is the rejected waste by human. It constitutes one of the greatest and dangerous wastes, which management still remains a great challenge worldwide. Tons of it are generated each day and its management varies from one region to another across the world. In advanced countries, fecal sludge is being valorized not only as biogas source but also as soil amendment after being stabilized via anaerobic digestion or co-composting with others organic materials [1-2]. As a consequence, the basic sanitation coverage rate is nearly 95% while many countries in developing world found difficulty to achieve 75% coverage; which is Millennium Development Goals for sanitation [3]. In 2014, it was reported that nearly 2.5 billion people were without enhanced sanitation, of which about 1 billion people practiced open defecation [3]. Further investigation by the same institution revealed that Sub-Saharan Africa, Oceania and Southern Asia, were the most vulnerable regions characterized by the lowest sanitation coverages (30%, 35% and 42%, respectively). Mostly in Africa, waste management represents a great challenge; the technology to encounter the challenges are often not
available at local level. Such situation lead to a chronic fecal-oral contamination, pathogen dissemination and water contamination; which represent one of the main factors responsible of the death of over 50% of child in the developing countries [3-5]. According to Bill and Melinda Gates Foundation [6], food and water contamination with fecal matter are the cause of diarrhea of over 2.5 billion children and the death of 600,000 children. The treatment and management of liquid and solid wastes has become one of the center of interest in scientific research. Otherwise, the search of the development of alternative renewable energy sources and to upgrade the conversion of organic materials into biogas keep growing [7].

Anaerobic digestion (AD) is a well-established process in which bacteria convert organic wastes to a methane and CO\textsubscript{2} gas mixture (generally about 60% methane and 40% CO\textsubscript{2}) called biogas and into a value added by-product known as biological fertilizer rich in nitrogen and phosphorous for the benefit of crops [8-9]. However, if many organic wastes can easily be converted to produce biogas and stabilized fertilizer, it is not the case with fecal sludge. Indeed, little is known about fecal sludge anaerobic digestion. This could be justified by the repulsive nature of this waste and the connotation or the view human has regarding fecal sludge. Snell [10] was the first to investigate on this issue, he reported a production of 0.5 m\textsuperscript{3} kg\textsuperscript{-1} VS during anaerobic digestion. The same author reported the inhibition of anaerobic digestion when mixing feces and urine as result of high concentration of nitrogen in the latter. Colón et al. [11-12] performed undiluted simulant human excreta in mesophilic condition; and reported that biogas yield ranged 0.24 to 0.44 NL biogas g\textsuperscript{-1} COD and COD removal efficiency was about 80%. Others studies [13-14] just focus on treating and removing COD from fecal sludge.

C: N ratio of fecal sludge is generally low due to the high concentration of nitrogen in this matter. This factor often limits anaerobic digestion process of the matter and leads to low biogas production. In addition, it has been reported that during anaerobic digestion of human excreta, there is high production of sulfuric acid (H\textsubscript{2}S) which is harmful to bacteria growth, limit biogas production and it is finally responsible of undesirable sickening odor [15]. Otherwise, it is known that that providing of electron donor in anaerobic reactor can improve the digestion process and biogas yield. To this end, iron zero valent (nZVI), micro iron pounder (IP), and iron scrap have been successfully used to enhance methane yield during various organic wastes [16-20], elimination of odors (such as H\textsubscript{2}S), and better sludge stabilization [15; 21] during anaerobic digestion process. However, despite the wide application of electron donors during anaerobic digestion, it application in fecal sludge digestion has not yet been documented.

Therefore the objective of this study is investigate the effect of micro iron powder during anaerobic digestion of fecal sludge. The effect of iron powder on methane gas production, COD removal and sludge stabilization for reuse as organic fertilizer in agriculture will be examined. To the best of our knowledge, this is the first report on the application of electron donor during fecal sludge anaerobic digestion.

2. Materials and Methods

2.1. Fecal Sludge and Micro Iron Powder

Fecal sludge used as a substrate in this study was a freshly deposited feces collected from a household. Feces was collected in a 500 mL glass bottle and brought to the laboratory. The sludge underwent pretreatment which consisted into the determination of physicochemical parameters such as pH, water content (W), dry matter (DM), organic matter (OM), total organic carbon (TOC), volatile matter (VM), chemical oxygen demand (COD) and the total Kjeldahl nitrogen (TKN) determination. Iron powder (purity > 98% and diameter 0.2 mm) was purchased from Sinopharm Chemical Reagent Co. Ltd.

2.2. Analytical Methods

The pH was measured in the fresh sample after being diluted to 15% total solids. A multi parameters HANNA COMBO was used. Water content, alkalinity, COD and OM were determined according to the American standard for the examination of water and waste water [22]. The total organic carbon (TOC) was determined from OM following the equation (1) [23]. TKN was determined according to French norm AFNOR NFT90-110.

\[
\text{TOC} = \frac{\text{OM}}{1.8}
\]

2.3. Experimental Setup for Biogas and Methane Measuring

The experimental setups was at first set to monitor the production of raw biogas and secondly to directly measure the volume of methane. The bioreactor design was inspired from study carried out by [24]. Briefly, the digester was consisted of a 1000 mL glass bottle. A hole was perforated on the cover of the bottle and through this orifice, the digester was connected via a PVC pipe to a calibrate glass cylinder containing tap water. The biogas produced in the digester passes through the connection pipe to reach the calibrated cylinder; under which it generate bubbles to ascend to the bottom and generate a pressure which displace the tape water. The displacement of water is equal the volume of biogas produced. The entire dispositive is shown schematically in Figure 1. The use of NaOH afterward permitted to absorb unwanted gases and trace gas such as carbon dioxide (CO\textsubscript{2}) and hydrogen sulfide (H\textsubscript{2}S) in order to directly determine the volume of methane generated.
2.4. Experimental Design

Four (4) batches experiments were set up. In these series of experiments, fecal sludge was enriched with iron powder at different concentrations: A (400 g sludge+0 g Fe), B (400 g sludge+0.5 g Fe) C (400 g sludge+1g Fe), and D (400 g sludge+3g Fe). The samples were well mixed and diluted with distilled water to 15% dry matter (DM) and homogenized by mechanical agitation [24-25]. No pH adjustment was proceeded as the later was in the range of neutrality, favorable condition for bacterial activity [26-27]. Digesters were loaded and kept in a water bath at 45°C (thermophile condition) for 17 days (Figure 2).

3. Results and Discussion

3.1. Physicochemical Parameters of the Raw Excreta

The main physicochemical parameters determined are shown in Table 1. Data showed that fecal sludge has a near neutral (pH 7.6) favorable for a good anaerobic digestion. Excreta have a high organic matter content (94.7% dry matter) highlighting the probable presence of high proportion of biodegradable matters. Fecal sludge also has a high COD concentration (2217 mg O₂ L⁻¹). The direct discharge of such material into the environmental without any pre-treatment would represent a threat to the ecosystem of the receiving environment. The C: N or TOC: TKN ratio is equal to 11.6. This value is low and doesn’t favor good anaerobic digestion, as it is reported in previous studies [28-29], during anaerobic digestion, microorganisms consume carbon 25-30 times faster than nitrogen. So therefore, for a good anaerobic digestion, it is recommended to keep the C: N ratio between 20: 1 and 30: 1. In this study, we didn’t correct the C: N ratio because we want to avoid adding any other organic material to feces; but we added electron donors (micro iron powder Fe) to catalyze the digestion as suggested by [18; 20; 24-25] for sewage sludge.

3.2. Optimization of Fecal Sludge Digestion: Effect of Iron Powder

3.2.1. Biogas Production

The evolution of biogas production, both daily and cumulative during anaerobic digestion is displayed in Figure 3a and 3b.

![Figure 1. Setup for produced biogas measurement.](image1)

![Figure 2. Image showing anaerobic digestion in progress.](image2)

<table>
<thead>
<tr>
<th>Parameters</th>
<th>W</th>
<th>DM</th>
<th>pH</th>
<th>OM</th>
<th>COD</th>
<th>TA</th>
<th>TOC</th>
<th>TKN</th>
<th>TOC/TKN</th>
</tr>
</thead>
<tbody>
<tr>
<td>Values</td>
<td>82.5</td>
<td>17.5</td>
<td>7.6</td>
<td>94.7</td>
<td>2217</td>
<td>653.8</td>
<td>52.6</td>
<td>4.6</td>
<td>11.4</td>
</tr>
</tbody>
</table>

Note: W, DM, OM, TOC and TKN are in (%), COD in (mg O₂ kg⁻¹) TA in mg CaCO₃ kg⁻¹.
Figure 3a shows the profile of the daily production of biogas in the different digesters. From the data, it can be observed low production of biogas as the consequence of low bacterial activity at the beginning of the experiment. In the digester A (control), it took 4 days for biogas to start being produced. In another word, it took 4 days for an intense activity of microorganisms to be noticed. This period of time is the acclimatization time during which microorganisms adapt to the material. After 4 days of acclimatization, a sudden increase in biogas production with a daily production up to 1000 mL kg$^{-1}$ sludge on the 5th day of digestion can be noticed. Biogas production was kept around 1000 mL kg$^{-1}$ till the 9th day and then increased slightly to 1200 mL kg$^{-1}$ sludge on the 10th day. From the 10th day biogas production dropped until the end of the process at the day 14. The sudden increase of the volume of biogas from the 4th day reflects an intense bacterial activity. Indeed, during anaerobic digestion, microorganisms use organic carbon as energy source for the renewal of their cells. As a consequence organic matter are degraded and convert into biogas [24]. Biogas production decrease would explain the reduction in bacterial population due to the death of certain microorganisms during the degradation of organic matter. The deficit of “food” causes the death of microorganisms provoking the fall of biogas production. Unlike to the control, the digesters B, C and D containing respectively 0.5 g; 1 g and 3 g of iron powder have a relatively short acclimatization period (2 days) and intense bacteria activity was noticed earlier. A remarkable biogas production was noted from the second day until the 12th day. As it can be seen, the duration of the anaerobic digestion was also shorten in those 3 digesters. The optimal biogas volumes obtained were respectively 1683 mL kg$^{-1}$ sludge, 1683 mL kg$^{-1}$ sludge and 1100 mL kg$^{-1}$ sludge respectively in the digesters B, C and D. Compared to the control (digester A), biogas production was remarkably improved in the digesters B and C. A comparison of biogas production shows that digesters containing iron powder (B, C, D) have a remarkable performance compared to the digester A. The total produced volume of biogas was estimated at 8315 mL kg$^{-1}$ sludge, 12000 mL kg$^{-1}$ sludge, 12800 mL kg$^{-1}$ sludge and 9535 mL kg$^{-1}$ sludge, respectively in the digesters A, B, C and D (Figure 3b). Results showed that the presence of iron powder in the digesters contributed to improve biogas production. Indeed, in a previous study on sewage sludge, it was proved that external supply of electron in an anaerobic digester enriched the environment and promoted better microorganisms’ activity [16; 18-19; 31]. In this case with concentrations of 0.5 and 1 g of iron, the digestion of fecal sludge showed better performance. Otherwise, it is also found that in the digester D containing 3 g of iron powder biogas production considerably diminished compared to the digester B and C (Figure 3b). The same
observation was noted by [30] then [24] in their study on the effect of zero valent iron nanoparticles (Fe⁰) and magnetite (Fe₃O₄) during anaerobic digestion of sewage sludge. It appeared that the concentration of iron powder which allows optimum biogas is comprised between 1.25 g and 2.5 g of Fe kg⁻¹ fecal sludge. However, though Fe is not an environment metals of concern, in order to avoid iron overload in the digested sludge and environmental iron contamination, minimizing its amount in anaerobic digester would be interesting and promising. This emphasize the need of more investigation combining fecal sludge with others organic materials possessing high C:N ratio.

3.2.2. Methane in the Biogas

The second experimentation which consisted in adsorbing unwanted biogas such as CO₂, in alkaline solution has permitted to directly measure the volume of methane gas. The result is shown in Figure 4a. One can clearly observe that methane production follows the same profile as that of biogas aforementioned. The effect of iron powder on methane production can quickly be noticed. Indeed, the presence of iron powder in the digesters B, C and D has significantly enhanced methane production compared to the digester A; which contained any trace of iron powder. The total volume of methane produced was: 4823 mL kg⁻¹ sludge, 8688.5 mL kg⁻¹ sludge, 9933 mL kg⁻¹ sludge and 7132 mL kg⁻¹ sludge, respectively in the digesters A, B, C and D (Figure 4b). It can thus be seen that compared to the control (digester A), the presence of iron powder has doubled the volume of methane in digesters B and C. The best performance is obtained in the digester C containing 1 g of Fe. This best performance of the anaerobic digestion and methane gas production in the presence of iron powder can be explained not only by the enrichment of bacterial activity in the environment due to the electron released in the biodigesters by the electron donors (Fe → Fe²⁺+2e or Fe → Fe³⁺+3e) [16; 31]; but also the imminent conversion of part of the CO₂ produced to CH₄ via the equation (2) [15; 18]. Consequently, the volume of produced methane increased while CO₂ production considerably reduced. Furthermore, it has been also reported that part of produced CO₂ could be converted into CH₄ by fixing H₂ (which could be produced during Fe corrosion) as showed in equation (3) [20]. The process of the reaction between CO₂ and H₂ could pass by an intermediary to methane, which will be further convert into methane (equation (4) and (5)) [18; 25]. Moreover, it has been given to us to notice that during the digestion, the unwanted odor caused by the formation of H₂S was greatly reduced as the sickening other decreased in the presence of iron powder. Indeed, Fe⁰ could remove toxic and biologically harmful compounds such as trace of H₂S by precipitating the produced S²⁻ via equation (6) and preventing the formation of

![Figure 4. Methane production in each digesters](image)

(a) daily volume and (b) cumulative volume.
H₂S responsible of the unwanted odor [15]. This would not only buffer the system and lead to more stable methane production but also better stabilized the sludge with less odor.

\[
8H^+ + 4Fe^0 + CO_2 \rightarrow CH_4 + 4Fe^{2+} + 2H_2O ; \Delta G^0 = -150.5 \text{ kJ mol}^{-1}
\]
\[
4H_2 + CO_2 \rightarrow CH_4 + 2H_2O ; \Delta G^0 = -131 \text{ kJ mol}^{-1}
\]
\[
4H_2 + 2CO_2 \rightarrow CH_3COO^- + H^+ + 2H_2O ; \Delta G^0 = -95 \text{ kJ mol}^{-1}
\]
\[
CH_3COO^- + H^+ \rightarrow CH_4 + CO_2, \Delta G^0 = -75.7 \text{ kJ mol}^{-1}
\]
\[
Fe + H_2S \rightarrow FeS + H_2
\]

3.2.3. Carbon Dioxide

The effect of iron powder on methane production has been highlighted by evaluating the total volume of CO₂ produced during the overall anaerobic digestion process in each digester (Figure 5). It can clearly be noted from the Figure that CO₂ volume diminished with the concentration of iron powder. This would explained the increase of methane yield as aforementioned.

3.2.4. Biogas Composition

Based on the aforementioned results regarding biogas and methane production, and on the fact that biogas generally consists of methane and carbon dioxide [8-9], the composition of the produced biogas in each bio-digester was determined and results are shown in the Table 2. According the data, the presence of iron powder in the digesters enhanced methane concentration in biogas while decreased CO₂ concentration. The highest percentage of methane achieved was 77.6% in the digester C containing 1g Fe. To be noticed, although high concentration of Fe has a negative impact on the performance of microorganisms and biogas production in the digester D, it did improve methane concentration.

<table>
<thead>
<tr>
<th>Digesters</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
</tr>
</thead>
<tbody>
<tr>
<td>% CH₄</td>
<td>58.0</td>
<td>72.5</td>
<td>77.6</td>
<td>74.8</td>
</tr>
<tr>
<td>% CO₂</td>
<td>42.0</td>
<td>27.5</td>
<td>22.4</td>
<td>25.2</td>
</tr>
</tbody>
</table>

3.3. COD

COD reduction is one of the main objectives to achieve during anaerobic digestion of organic wastes. Figure 6 displays the removal of COD during anaerobic digestion. The initial concentration of chemical oxygen demand in the fecal sludge was 2217 mg O₂ L⁻¹. In the current work, the presence of iron powder in the bioreactor significantly improved the COD abatement. Indeed, COD decreased from 2217 mg L⁻¹ in the original fecal sludge to 1280.4; 885.4; 765.4 and 1097 mg L⁻¹, respectively in the digester A, B, C and D, at the end of the digestion. Those values correspond respectively to a 42.2%, 60.1%, 65.5%, 50.5% removal efficiency. The improvement of COD abatement in the presence of iron powder could be explained on one hand by the reduction power of Fe and by the anaerobic corrosion which undergoes Fe [32-33] on the other hand. According to the authors, during the anaerobic digestion, iron corrodes and an oxyhydroxide layer is forms on the surface of [32]. The oxyhydroxide layer formed provides sites for the adsorption of organic and inorganic pollutants [33]; which is reflected by the decrease of their concentration in the liquid phase of the digestate.

\[
Fe^0 + 2H_2O \rightarrow FeOOH + 1.5 H_2
\]

4. Conclusion

This study allowed us to appreciate the role of electron donors (Fe) on fecal sludge valorization during anaerobic digestion process. It appeared from this study that the anaerobic digestion of fecal sludge alone does not yield good methane and digestate is not well stabilized with remarkable nauseating odor. But when adding iron powder is added in a reasonable amount (1.25 to 2.5 g kg⁻¹ wet sludge), the process could be improved with a very good methane yield.
achievement. In addition, the abatement of COD was improved by 15% with the application of electron donor in anaerobic digester. Otherwise, the digested sludge was well stabilized with less odor, for further land application. Indeed, Fe could remove toxic and biologically harmful compounds such as trace of H₂S by precipitating the produced S²⁻ (Fe + H₂S → FeS + H₂) and preventing the formation of H₂S responsible of the unwanted odor. However, though Fe is not an environmental metals of concern, in order to avoid environment iron contamination, reducing or minimizing its amount in anaerobic digester would be interesting and promising. Therefore, we are looking forward to investigating the co-digestion of fecal sludge and others organic materials with high C:N ratio in the presence of iron powder.

References


