Biogas Recovery from Sewage Sludge during Anaerobic Digestion Process: Effect of Iron powder on Methane yield

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Abstract

The management of sewage sludge remains a great challenge in developing countries; as it hinder the development mainly in the big towns. Here we have conducted the anaerobic digestion experiment for recovering methane gas from sewage sludge. Iron power (IP) was applied and its impact of methane yield was investigated. Results showed that sewage sludge is a reservoir of energy in the form of methane gas. Methane recovery was greatly improved by adding IP in the AD reactor, as up to 141917.5 mL kg⁻¹ VS could be recovered when IP is properly used compared to 98783.4 mL kg⁻¹ VS in the blank. More specifically, methane yield was upgraded by 9.2%, 28.6% and 43.6% respectively at the dose rate of 0.3 g IP, 1 g IP and 3 g IP in 300 g of sludge (wet weight). Results also show that over dose concentration of IP (addition of 6 g IP) exercises a strong negative impact on AD process and methane yield.

Keywords: Sewage sludge, Anaerobic digestion, Iron powder, Biogas recovery.

Introduction

Progress, development and consumption are among the words that better characterize our current life. However, improving our lifestyle through the industrialization of our society has created pollution; that weakens and destroys our living environment. Nowadays, huge amount of municipal solid and liquid wastes are generated in modern societies; and its disposal poses serious environmental, social and economic problem which hinders development in developing countries, especially in the big cities. Solid waste management has become one of the major problems the world is facing today. The rapid increase in the generation of huge quantity of wastes is one aspect of the environmental crisis. According to Mane et al, annually organic waste generates from cites in India is nearly 700 million tons; while Chu et al reported over 11.2 million tons in China. In the poorest countries and mainly in Africa, there is generation of tones of wastes per days including; fruits and vegetable wastes and mainly human excreta; which management still remains a great challenge. The need of energy is global in our today’s life. The demand of energy is steadily increasing that the need for exploring and exploiting renewable, sustainable as well as eco-friendly sources is inevitable, for the fossil-based fuels has become scarce and more expensive.

Anaerobic digestion (AD) or methane fermentation is an economical and eco-friend process for biomass, organic matter conversion to produce biogas; which mainly consists of methane and carbon dioxide. It is a biological conversion of complex substrates into biogas and inert digestate by microbial activity in oxygen free environment. The digestion process involves four main steps, namely hydrolysis, acidogenesis, acetogenesis and methanogenesis. In advanced countries, wastes, wastewater, are recycled via anaerobic digestion process to produce biogas and an inert product named sewage sludge usable in agriculture or land applied. This technology is not yet well known and established in developing countries as it is the case in Africa.

Therefore, the present study aims to investigate the opportunity of recovering biogas (methane) from sludge and improve methane yield. To the best of our knowledge, this would be a first investigation in the republic of Benin.

Materials and Methods

Raw material and experimental design: Sewage sludge used as substrate in this study was collected from municipal waste treatment plant (WWTP). Sludge sample was characterized before use (Table-1). A total of five (5) sets of experiments were setup each in triplicate. Sludge was spiked with iron powder (IP) at different concentrations: A (300 g sludge + 0 g IP), B (300 g sludge + 0.3 g IP), C (300 g sludge + 1 g IP), D (300 g sludge + 3 g IP) and E (300 g sludge + 6 g IP). Samples were well mixed, and diluted by adding distilled water to obtain 15%...
of total solid (TS) then homogenized by mechanical shaking. No pH adjustment was made as the pH (6.8) of the mixtures is already closer to the neutrality; which is the favorable pH for better bacteria activity\(^1\). 500 mL glass bottles were used as anaerobic digesters. After being fed, each bottle was flushed with nitrogen gas for about three (3) minutes to maintain anaerobic condition inside the digester. Bottles were then kept in thermophilic condition into the water bath at 45°C. Each sample was in triplicate.

**Biogas measurement and composition:** The digester was connected to a calibrate glass cylinder containing tap water (Figure-1). The volume of biogas was measured as the displacement of water within the calibrate glass cylinder. Gas samples were periodically taken and its composition (methane and carbon dioxide) were analyzed using gas chromatography. Gas standards consisting of 100% CO\(_2\) and 100% CH\(_4\) were used.

Physicochemical parameters include total solid (TS), volatile solids (VS), Total organic matter and total alkalinity (TA) were conducted in accordance with Standard Methods for the Examination of Water and Wastewater\(^2\). pH was determined using a multi-parameter (type HACH, HQ40d). Total organic carbon (TOC) was determined according to Jiménez and gracia\(^3\) (equation-1). Total Kjeldahl Nitrogen (TKN) was determined following standard method AFNOR NF190-110. C/N ratio was calculated as the ratio of the percentage of TOC to the TKN. Chemical oxygen demand (COD) and TA, were determined in the supernatant of the dissolved sample in the distilled water after being centrifuged at 5000 x g for 15 min at 4°C.

\[
TOC = \frac{\text{TOM}}{1.8} \tag{1}
\]

**Table-1**

<table>
<thead>
<tr>
<th>Samples</th>
<th>pH(_i)</th>
<th>pH(_f)</th>
<th>TA(_i) (mg L(^{-1}))</th>
<th>TA(_f) (mg L(^{-1}))</th>
<th>COD(_i) (mg L(^{-1}))</th>
<th>COD(_f) (mg L(^{-1}))</th>
<th>TS(_i) (%)</th>
<th>C/Ni</th>
</tr>
</thead>
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<td>Sludge</td>
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<td>-</td>
<td>886.4</td>
<td>3219</td>
<td>3219</td>
<td>3219</td>
<td>23.03</td>
<td>11.86</td>
</tr>
<tr>
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<td>7.7</td>
<td>886.4</td>
<td>987</td>
<td>3219</td>
<td>1456.7</td>
<td>15</td>
<td>11.86</td>
</tr>
<tr>
<td>B</td>
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<td>7.3</td>
<td>886.4</td>
<td>995</td>
<td>3219</td>
<td>1375.6</td>
<td>15</td>
<td>11.86</td>
</tr>
<tr>
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<td>7.5</td>
<td>886.4</td>
<td>978</td>
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<td>1126.8</td>
<td>15</td>
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</tr>
<tr>
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<td>7.2</td>
<td>886.4</td>
<td>917</td>
<td>3219</td>
<td>946.5</td>
<td>15</td>
<td>11.86</td>
</tr>
<tr>
<td>E</td>
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<td>7.4</td>
<td>886.4</td>
<td>937</td>
<td>3219</td>
<td>1570.9</td>
<td>15</td>
<td>11.86</td>
</tr>
</tbody>
</table>

\(^i\) = initial and \(^f\) = final

**Figure-1**

Results and Discussion

Biogas and methane production: Biogas production is a key parameter to appreciate the anaerobic digestion process. The quantity and quality of biogas, is often used to appreciate how good the AD process is. In this study, wet biogas production was followed during the experiment. Daily and cumulative volume of wet biogas, methane is respectively shown in the figures-2a and figures-2b. From the figure-2a, it can be seen that biogas generation started right on the day 1 of the experiment in all digesters. Daily and highest biogas production was achieved on the day 6 in all digesters: A (33748 mL kg\(^{-1}\) VS), B (35230 mL kg\(^{-1}\) VS), C (44120 mL kg\(^{-1}\) VS), D (39510 mL kg\(^{-1}\) VS) and E (16298 mL kg\(^{-1}\) VS). The high biogas production is the result of intensive bacteria activity within AD bioreactors. From the day 6\(^{th}\), biogas production rate decreased as result of diminution of microbes’ population and activities. By the end of the process, after 12\(^{th}\) of digestion, no more biogas was produced in all digesters; indicating no more bacteria activity. Results show that biogas production is IP dependent. Biogas yield increased as IP concentration increased. However, high concentration of IP exhibits a strong negative impact on bacteria activity and biogas yield\(^{8}\) like it is the case in the digester E. The shortest duration of the digestion process (12 days) compared to the often reported duration in literature (up to 30 days) could be explained on one hand by the high microbial activity stimulated by the addition of IP; and on the other hand due the fact that the used raw sludge was freshly collected and rich in anaerobic bacteria. At the end of the digestion, an average volume of biogas generated was evaluated to 160181, 160510, 179771, 192118 and 121494 mL kg\(^{-1}\) VS respectively in the digester A, B, C, D, and E (figure-2b). Base on wet biogas production, the efficiency of anaerobic fermentation can be classed as follow: E < A < B < C < D.

![Figure-2](image-url)

Figure-2
Biogas production during anaerobic digestion (a) daily volume and (b) cumulative volume
The main objective during AD is to recover methane gas which could serve as combustible in various area of the economy, including generation of electricity, heating, and in kitchen. Beyond wet biogas, methane is the key factor for better appreciating AD process. In order to evaluate the volume of methane produced, wet biogas content was determined twice during the experiment, and the average methane content is as followed: A (61.6%), B (67.2%), C (70.7%), D (73.8%) and E (71.3). As results, daily and cumulative volumes of produced methane are estimated as shown in the figures-3a and figures-3b. Methane production curve showed similar pattern with wet biogas (figure-3a). By the end of the digestion, methane yield was evaluated to: A (98783.4 mL kg\(^{-1}\) VS), B (107910.7 mL kg\(^{-1}\) VS), C (127080.2 mL kg\(^{-1}\) VS), D (141917.5 mL kg\(^{-1}\) VS) and E (86649.2 mL kg\(^{-1}\) VS) (figure-3b). As a consequence, the efficiency of AD process base on methane yield follows the order previously mentioned with wet biogas.

**Impact of IP on AD process, COD removal and methane yield upgrading:** Iron powder also known as zero valent iron owing to its reducing property and their capability to produced electrons (Fe → Fe(2+) + 2e), greatly enhance anaerobic fermentation process. Indeed, when comparing biogas production in the digesters with IP with control (digestor A), it can clearly be seen that methane yield was improved by 9.2%, 28.6% and 43.6%, respectively in the digester B (+ 0.3 g IP), C (+ 1 g IP) and D (+ 3 g IP) (figure-4). Such enhancement of methane yield could be explained by the enrichment of the medium within the bioreactor in the presence of IP. The improvement of methane yield is function of IP concentration; and it increases with the increase of IP concentration\(^6\).
Nevertheless, when the concentration of IP is too high, beyond certain limit, it exhibits a strong negative impact on bacteria activity and methane production as it is the case in the digester E at 6 g IP where methane yield decreased by 12.3%. Yang et al. reported the accumulation of hydrogen in the presence of zero valent iron at high concentration. Such accumulation of Hydrogen within the reactor would increase the partial pressure of hydrogen; which thermodynamically does not favor methane forming bacteria activity. The effect IP on methane yield enhancement could better be explained when having a look on CO₂ volume generated in each digester (figure-5). Results show that the volume of produced carbon dioxide decreased in the presence of IP. Its diminution is also function of IP concentration. The total volume of carbon dioxide produced in the digester A (control) is estimated to 56591.8 mL kg⁻¹ VS compared to 49388.9 mL kg⁻¹ VS, 47297.7 mL kg⁻¹ VS and 30988.6 mL kg⁻¹ VS and 36059.3 mL kg⁻¹ VS, respectively in the digester B (+ 0.3 g IP), C (+ 1 g IP), D (+ 3 g IP) and E (+ 6 g IP). This is due to the conversion of part of the produced CO₂ to methane via the equation-2 below; updating CH₄ yield and lowering CO₂ volume.

\[
8\text{H}^+ + 4\text{Fe}^{0} + \text{CO}_2 \rightarrow \text{CH}_4 + 4\text{Fe}^{2+} + 2\text{H}_2\text{O} \Delta G_0 = -150.5 \text{ KJ/mol CH}_4
\]  

(2)
The presence of IP in the AD bioreactor also improved sludge’s COD removal. COD decreased from 3219 mg L\(^{-1}\) in the raw sludge before AD to 1456.7, 1375.6, 1126.8, 946.5 and 1570.5 mg/L, respectively in the digester A, B, C, D and E (table-1). These respectively correspond to the removal efficiency of 54.7%, 57.3%, 65%, 70.6% and 51.2%. Indeed, in previous study\(^{15}\) it has been reported that under anaerobic digestion as it is the case in the present study, iron zero valent reacts with water and forms oxyhydroxide layer on the particle’s surface as described below (equation-3)\(^{15}\). The formed oxide shell provides sites for sorption of pollutants such as chemical organic compounds; which results in the diminution of their concentration in the liquid phase of the sludge\(^{16}\).

\[
\text{Fe}(0) + \text{H}_2\text{O} \rightarrow \text{FeOOH} + 1.5 \text{H}_2 \text{ (Anaerobic corrosion)} \quad (3)
\]

**Conclusion**

Methane recovery from sewage sludge during AD was studied and the effect of iron powder on methane production was investigated. Results show that sludge is a potential reservoir for energy (methane) which could be valorized. The utilization of iron powder could also help to better and efficiently recover methane from sludge. However, overdose of iron powder will inhibit AD process and methane yield.

**References**