Pretreatment followed by anaerobic digestion of secondary sludge for reduction of sewage sludge volume
Naoki Abe, Yue-Qin Tang, Makoto Iwamura, Shigeru Morimura and Kenji Kida

ABSTRACT

The influence of two pretreatment methods, thermal treatment and low-pressure wet oxidation, on the sludge digestion efficiency was examined. Batch thermophilic anaerobic digestion was used to evaluate the effectiveness of the pretreatment methods in terms of volatile suspended solids (VSS) digestion efficiency and gas production. The results showed that the gas production was not proportional to the VSS degradation efficiency of either thermal treatment or low-pressure wet oxidation. Low-pressure wet oxidation treatment at 150 °C along with 40% of the theoretical oxygen required to oxidize organic carbon gave the highest gas production and the VSS digestion efficiency of 77% at a VSS loading rate of 8 g l⁻¹ d⁻¹. The digestion efficiency was about 30% higher than that of thermophilic anaerobic digestion without sludge pretreatment. Sewage sludge could be treated effectively at a high VSS digestion efficiency with this pretreatment followed by thermophilic anaerobic digestion.

INTRODUCTION

As economic activity is increasing worldwide, the number of sewage treatment plants is increasing, thus resulting in a rapid rise in the amount of sewage sludge produced. In Japan, more than 2,000,000 tons of dried sewage sludge is currently generated every year (Shirasaki & Yamaguchi 2011).

The sludge volume reduction process is the process of decomposing organic substances in order to reduce space for final disposal. One typical form of sludge volume reduction is incineration. In Japan, at present, about 70% of sludge was incinerated after being thickened and dewatered (Japan Sewage Works Association 2007). However, incineration produces dioxins and furans because of incomplete combustion and is energy inefficient (Sakamoto et al. 2001). Future increase in sludge generation is unavoidable. Therefore, usage of sewage sludge as a carbon-neutral energy source needs to be promoted (Tsuno & Kusuda 2011). Anaerobic digestion is commonly known to be an effective treatment for sewage that results in the reduction of sludge volume and the production of energy-rich biogas. Life-cycle assessment of sewage sludge treatment shows that anaerobic digestion leads to the lowest economic and environmental impacts owing to the significant reduction of sludge volume (Hong et al. 2009). However, the application of anaerobic digestion is limited because of its relatively long retention time and low digestion efficiency, which are generally because of the difficulty of the hydrolysis of sludge (Appels et al. 2008). Various pretreatment techniques such as ultrasonic cavitation (Erden & Filibeli 2009), ozonation (Bougrier et al. 2006), peracetic acid oxidation (Appels et al. 2011), high-pressure homogenization (Stephenson et al. 2007), and thermal treatment (Li & Noike 1992; Bougrier et al. 2008; Morgan-Sagastume et al. 2011) have been studied to improve the biogas production efficiency by enhancing the hydrolysis of secondary sludge (Appels et al. 2008). However, these pretreatment techniques, except for thermal treatment, have not yet been applied owing to their high costs (Kalogo & Monteith 2001). A few thermal hydrolysis processes have been commercialized and are operated at full-scale as the Cambi and Biothelys processes. The Cambi thermal hydrolysis process, which treats sludge at 165 °C for 30 min (Kepp et al. ...
was first installed in 1995 at Hamar, Norway. The Biothelys process, similar to the Cambi thermal hydrolysis process, was commercialized by Veolia Water. This process was first installed in 2006 in France (Chauzy et al. 2008). Now, over 20 plants are being operated in Europe and some are planned worldwide (Tsang & Sapienza 2011). However, the maximum digestion efficiency is approximately 65% in these commercialized applications of the Cambi and Biothelys processes (Panter & Kleiven 2005; Chauzy et al. 2008).

We have previously developed a high-efficiency anaerobic digestion process for sewage sludge using a modified two-stage digestion process. The thickened secondary sludge is initially liquefied in thermophilic anaerobic digestion at 53°C and then mixed with primary sludge, after which the mixture is treated in a mesophilic anaerobic digester at 37°C. Compared with conventional methods, the maximum volumetric loading rate of volatile suspended solids (VSS) achieved in the modified two-stage process was two to three times higher, but VSS anaerobic digestion efficiency was only 10% higher at the same volumetric loading rate (Kida & Ikbal 1995).

In the present study, to further improve VSS digestion efficiency, the thermal treatment commercialized as described above was firstly evaluated. A low-pressure wet oxidation process was then studied as moderate pretreatment. The thermophilic batch anaerobic digestion of the pretreated thickened secondary sludge was conducted to evaluate the effect of pretreatment on gas production efficiency. Moreover, the thickened secondary sludge treated by the low-pressure wet oxidation was subjected to the treatment test of thermophilic anaerobic digestion, using a continuous stirred tank reactor (CSTR).

**METHODS**

**Sludge characteristics**

Sludge samples were provided by Kumamoto Northern Sewage Treatment Plant (Kumamoto City, Japan). Thickened secondary sludge for the pretreatment study was obtained from a sludge buffer tank installed behind the decanter centrifuge. Seeding sludge for anaerobic digestion was acclimated using a thermophilic anaerobic digester with agitator. Table 1 shows the composition of the thickened secondary sludge used in the experiments.

**Table 1** Typical compositions of thickened secondary sludge used through experiments

<table>
<thead>
<tr>
<th>General property</th>
<th>For thermal treatment</th>
<th>For low-pressure oxidation</th>
<th>For anaerobic digestion*</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS (mg l⁻¹)</td>
<td>40,590</td>
<td>32,250</td>
<td>41,030</td>
</tr>
<tr>
<td>VTS (mg l⁻¹)</td>
<td>32,670</td>
<td>28,060</td>
<td>36,850</td>
</tr>
<tr>
<td>SS (mg l⁻¹)</td>
<td>38,270</td>
<td>31,930</td>
<td>40,420</td>
</tr>
<tr>
<td>VSS (mg l⁻¹)</td>
<td>31,860</td>
<td>27,090</td>
<td>35,670</td>
</tr>
<tr>
<td>S-TOC (mg l⁻¹)</td>
<td>469</td>
<td>834</td>
<td>51.4</td>
</tr>
<tr>
<td>VFAs (mg l⁻¹)</td>
<td>499</td>
<td>1,538</td>
<td>ND</td>
</tr>
<tr>
<td>pH (–)</td>
<td>6.0</td>
<td>6.1</td>
<td>6.6</td>
</tr>
<tr>
<td>Color intensity ( )</td>
<td>172</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

TS: total solids; VTS: volatile total solids; SS: suspended solids; S-TOC: soluble total organic carbon; VFAs: volatile fatty acids.

*Used for the low-pressure oxidation followed by thermal anaerobic digestion.

**Thermal pretreatment**

A total of 1.0 L of the thickened secondary sludge was placed in the autoclave (volume 2 L, model TAS-2, Taiatsu Techno, Japan) as described previously (Zhu et al. 2004). The treatment was conducted at 120, 150, 170 and 200°C at 237 rpm for 1 h, and the autoclave was cooled rapidly by placing it in cold water.

**Low-pressure wet oxidation pretreatment**

The treatment was performed with the same experimental apparatus as the thermal treatment. After a total of 0.4 L of the thickened secondary sludge was placed in the autoclave, oxygen was supplied through a mass flow meter (model SEF-1420, STEC, Japan) at 0–120% of the theoretical amount of oxygen (13.6 g-O₂) necessary for oxidation of 5.11 g-carbon contained in 13 g-sludge. Treatment was conducted at 150°C, 0.5–1.4 MPa and 237 rpm for 2 h, and the autoclave was cooled rapidly by placing it in cold water.

The VSS degradation efficiency of the thickened secondary sludge in the pretreatments was calculated using the following equation.

\[
\text{VSS degradation efficiency} (\%) = \left(1 - \frac{V_f}{V_0}\right) \times 100
\]

\(V_0\): VSS concentration of the thickened secondary sludge (mg l⁻¹); \(V_f\): VSS concentration after pretreatment (mg l⁻¹).

**Gas production test by batch anaerobic digestion**

An aliquot of 350 ml of the seeding sludge and 50 ml of the pretreated secondary sludge were placed into a bottle...
working volume 400 ml). Bottles were sealed with rubber stoppers and were placed in a temperature-controlled water bath (53 °C). The liquid was stirred continuously with a bar magnet. The produced gas was then collected using a gas holder (Tanemura et al. 1994).

Anaerobic digestion of pretreated thickened secondary sludge

Anaerobic digestion of the pretreated sludge was conducted using an 8 L CSTR made of glass. The temperature of the reactor was maintained at approximately 53 °C and pH was not controlled. The produced gas was collected with a gas holder and the volume was recorded every day. Thermophilic anaerobic digestion sludge provided by Kumamoto Northern Sewage Treatment Plant was used as seeds for starting up the reactor. The thickened secondary sludge (see Table 1) was supplied once a day at a volatile total solids (VTS) loading rate of 3 g l\(^{-1}\) d\(^{-1}\) for about 2 months using the draw-and-fill method. Based on previous data (Kida & Ikbal 1998), the VTS loading rate was increased up to 8 g l\(^{-1}\) d\(^{-1}\) and the reactor was operated for 7 d. Then the pretreated thickened secondary sludge was added in the same manner for 15 d. The VSS digestion efficiency of the thickened secondary sludge with or without pretreatment was calculated using the equation of the degradation efficiency, in which \(V_0\) is VSS concentration of the thickened secondary sludge (mg l\(^{-1}\)); \(V_1\) is VSS concentration of the effluent from CSTR (mg l\(^{-1}\)).

Analytical methods

Total solids (TS), VTS, suspended solids (SS), VSS, NH\(_4^+\)-N, soluble total organic carbon (S-TOC) and total carbon (TC) were analyzed according to the standard methods (Japanese Industrial Standards Committee 1986). S-TOC was measured using a TOC analyzer (TOC-500, Shimadzu, Japan). The carbon content of dry samples was determined using a CHN coder (MT-3, Yanako, Japan). Volatile fatty acids (VFAs) were analyzed by the post-labeling method using a high-performance liquid chromatograph (880-PU, 860-CO, Japan Spectroscopic, Japan), as described previously (Kida et al. 1991). Color intensity was measured at 620 nm with a spectrophotometer (UV-160A, Shimadzu, Japan) using a standard solution consisting of K\(_2\)PtCl\(_6\) and CoCl\(_2\)-6H\(_2\)O.

RESULTS AND DISCUSSION

Thermal treatment of secondary sludge

Table 2 shows the effect of treatment temperature on the VSS degradation efficiency, S-TOC, and VFAs during the thermal treatment of the secondary sludge. The VSS degradation efficiency and S-TOC increased with increasing treatment temperature. In particular, the VSS degradation efficiency increased from 16% at 120 °C to 55% at 170 °C and 70% at 200 °C. It was found that treatment at higher temperatures increased the amount of S-TOC and achieved good VSS degradation efficiencies. The amount of VFAs

<table>
<thead>
<tr>
<th>Pretreatment</th>
<th>VSS (mg l(^{-1}))</th>
<th>S-TOC (mg l(^{-1}))</th>
<th>VFAs (mg l(^{-1}))</th>
<th>TS (g l(^{-1}))</th>
<th>C content (%)</th>
<th>Total C (g l(^{-1}))</th>
<th>Recovery efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Thermal treatment</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>No treatment</td>
<td>31,860a</td>
<td>469a</td>
<td>499a</td>
<td>40.59</td>
<td>38.89</td>
<td>15.79</td>
<td></td>
</tr>
<tr>
<td>Temp. (°C)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>120</td>
<td>26,840</td>
<td>5,105</td>
<td>1,317</td>
<td>39.80</td>
<td>39.53</td>
<td>15.73</td>
<td>99.6</td>
</tr>
<tr>
<td>150</td>
<td>20,550</td>
<td>7,065</td>
<td>1,409</td>
<td>40.01</td>
<td>39.82</td>
<td>15.93</td>
<td>100.9</td>
</tr>
<tr>
<td>170</td>
<td>14,350</td>
<td>9,635</td>
<td>1,623</td>
<td>38.31</td>
<td>39.04</td>
<td>14.96</td>
<td>94.7</td>
</tr>
<tr>
<td>200</td>
<td>9,440</td>
<td>10,710</td>
<td>2,935</td>
<td>33.51</td>
<td>39.71</td>
<td>13.31</td>
<td>84.3</td>
</tr>
<tr>
<td><strong>Low-pressure wet oxidation treatment</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>No treatment</td>
<td>27,090a</td>
<td>834a</td>
<td>1,538a</td>
<td>32.25</td>
<td>39.58</td>
<td>12.76</td>
<td></td>
</tr>
<tr>
<td>Oxygen (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>15,070</td>
<td>683</td>
<td>2,204</td>
<td>35.64</td>
<td>38.56</td>
<td>13.74</td>
<td>107.7</td>
</tr>
<tr>
<td>40</td>
<td>13,880</td>
<td>7,570</td>
<td>2,178</td>
<td>36.30</td>
<td>37.71</td>
<td>13.69</td>
<td>107.5</td>
</tr>
<tr>
<td>80</td>
<td>11,210</td>
<td>8,360</td>
<td>2,283</td>
<td>34.62</td>
<td>36.82</td>
<td>12.75</td>
<td>99.9</td>
</tr>
<tr>
<td>120</td>
<td>9,850</td>
<td>9,535</td>
<td>2,383</td>
<td>34.89</td>
<td>36.20</td>
<td>12.63</td>
<td>99.0</td>
</tr>
</tbody>
</table>

aVSS, S-TOC and VFAs of the thickened secondary sludge were the same as described in Table 1.
doubled at 200 °C, and the color intensity also increased from 1,075 W at 120 °C to 8,917 W at 200 °C and the color changed from brown to dark brown.

Low-pressure wet oxidation treatment of secondary sludge

Table 2 shows the effect of additional oxygen on the VSS degradation efficiency and S-TOC during low-pressure wet oxidation treatment. The VSS degradation efficiency and S-TOC increased with increasing oxygen volume. In particular, the VSS degradation efficiency increased from 49% at 40% (0.8 MPa) to 59% at 80% (1.0 MPa) and 64% at 120% (1.4 MPa) of the theoretical oxygen supply. However, the concentration of VFAs was almost constant under all conditions.

Gas production test with batch anaerobic digestion

Gas production efficiency was assessed by means of batch anaerobic digestion using different pretreated sludge and a non-pretreated sludge as a control. Gas production efficiency of the pretreated sludge was compared with that of the control during the first 7 d.

Figure 1(a) shows the amount of gas produced from the thermally treated sludge. The amount of gas produced increased with the temperature between 120 and 170 °C, with nearly 500 ml produced at 170 °C; for the control treatment, around 425 ml of gas was produced. This indicates a volume increase of about 18% through thermal treatment, probably due to the degradation of the cell wall of bacteria, which is the main component of secondary sludge. However, at 200 °C, only 335 ml of gas was produced, i.e., about 33% decrease in comparison with the amount of gas at 170 °C. Gas production was not proportional to the VSS degradation efficiency. TC was measured to conduct the mass balance of the thermal treatment process. As shown in Table 2, the recovery efficiency of TC at 200 °C decreased about 10% in comparison with the efficiency at 170 °C. The difference in the TC recovery efficiency suggested that high temperatures (~200 °C) lead to the production of recalcitrant soluble organics which cause the resulting dark brown liquor and/or toxic compounds. This phenomenon of soluble organics production has also been reported by other researchers (Delgenes et al. 2000; Dwyer et al. 2008; Wilson & Novak 2009).

Figure 1(b) shows the amount of gas produced from the low-pressure wet oxidation treated sludge. The amounts of gas produced at 0, 80 and 120% of the theoretical oxygen supply were similar (360–415 ml), whereas for the control treatment it was around 350 ml, indicating that the treatment at these oxygen levels increased the amount of gas by 3–19%. The amount of gas produced at 40% of the theoretical oxygen supply was 630 ml, approximately 1.8 times as much as with the control treatment. The gas production of sludge after low pressure wet oxidization was 360–415 ml (Figure 1(b)), which was approximately 85% of 460 ml of the gas production of thermal treatment sludge (Figure 1(a): temperature 150 °C). It was considered that the difference in the quantity of gas production was due to the difference in concentrations of VTS (28.1/32.7 × 100 = 85.9) of thickened secondary sludge used for the pretreatment. However, since only the quantity of produced gas for the system using sludge pretreated with 40% oxygen increased sharply, to exclude the possibility of experiment error the experiment was carried out once again using sludge pretreated with 40% oxygen. To confirm the result at 40% of the theoretical oxygen supply, the gas production test was carried out once again and the almost same result (closed triangle) was obtained. Gas production was also
not proportional to the VSS degradation efficiency although the TC recovery efficiency did not decrease at various conditions (Table 2). Thus, it is suggested that too much oxygen leads to the production of recalcitrant soluble organics and/or toxic compounds, hence reducing the gas production. The effect of operation pressure was studied at 150 °C and various pressures ranging from 0.4 to 1.4 MPa by supplying N2 into the autoclave instead of O2. Pretreated sludge was subjected to a batch anaerobic digestion test in the same manner. However, a difference in gas production was not obvious (data not shown).

For thermal treatment and low-pressure wet oxidation, the VSS degradation efficiency, S-TOC, VFAs as well as gas production efficiency evaluated through the batch anaerobic digestion of the pretreated sludge were compared (Table 2, Figure 1(a), (b)). The S-TOC and VFAs of sludge after thermal treatment and low-pressure wet oxidation treatment showed substantial hydrolysis. Thermal treatment featured the highest VSS degradation efficiency at 70.5%. Regarding gas production by batch anaerobic digestion, low-pressure wet oxidation treatment produced around 1.3 times as much gas as thermal pretreatment when the amount of theoretical oxygen supply in low-pressure wet oxidation was set at 40%. Therefore, low-pressure wet oxidation was much more suitable than thermal treatment for sludge pretreatment.

**Thermophilic anaerobic digestion of pretreated secondary sludge**

Thermophilic anaerobic digestion of the secondary sludge pretreated by low-pressure wet oxidation (0.8 MPa, 150 °C and a supplied oxygen amount of 40% of the theoretical oxygen) was studied using the CSTR. Figure 2 shows changes of the VSS digestion efficiency, gas production efficiency and pH during the thermophilic anaerobic digestion (53 °C, VTS loading rate 8 g l\(^{-1}\) d\(^{-1}\)) of secondary sludge pretreated. After the non-treated sludge was anaerobically treated for the 7 d, the pretreated sludge was fed into the CSTR and continued at the same VTS loading rate of 8 g l\(^{-1}\) d\(^{-1}\). The VSS digestion efficiency increased up to 77% with the operation time and the gas production efficiency also increased from 2,500 to 4,500 ml l\(^{-1}\) d\(^{-1}\), which was coincident with the result of the gas production test. This digestion efficiency was about 18% higher than that of the Cambi and Biothelys processes (Panter & Kleiven 2008; Chauzy et al. 2008), and also about 30% higher than that of thermophilic anaerobic digestion using un-pretreated sludge.

These results indicate that low-pressure wet oxidation treatment of the secondary sludge may overcome the limiting factors of the hydrolysis stage of sludge digestion. Adding this pretreatment to the previously developed modified two-stage digestion process (Kida & Ikbal 1995) would enable efficient digestion. The secondary sludge is initially liquefied by low-pressure wet oxidation and subsequently treated in a thermophilic anaerobic digester at 53 °C, and then mixed with the primary sludge. Finally, the mixture is treated in a mesophilic anaerobic digester at 37 °C. The VSS digestion efficiency of pretreatment followed by the modified two-stage digestion of the secondary sludge should be higher than that of the conventional digestion, and the gas production could be improved significantly.

![Figure 2](https://iwaponline.com/wst/article-pdf/67/11/2527/440326/2527.pdf)
CONCLUSION

Two pretreatment techniques were evaluated from the viewpoint of improving gas production efficiency of the secondary sludge. VSS degradation efficiency increased with treatment temperature in thermal treatment and increasing oxygen supply in low-pressure wet oxidation treatment. However, gas production was not proportional to VSS degradation efficiency for both pretreatments, suggesting that certain pretreatment conditions lead to the production of recalcitrant soluble organics and/or toxic compounds, and the gas production was reduced. Low-pressure wet oxidation with 40% of the theoretical oxygen necessary for the oxidation of carbon contained in the sludge, followed by anaerobic digestion, showed the highest gas production. VSS digestion efficiency at a VSS loading rate of 8 g·L⁻¹·d⁻¹ was at least 78%, which was about 30% higher than that of sludge without pretreatment.

REFERENCES


by thermal hydrolytic pretreatment. Water Res. 43, 4489–4498.

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