Application of vibration milling for advanced wastewater treatment and excess sludge reduction
Akira Sano, Akira Senga, Hiroshi Yamazaki, Hiroki Inoue, Kai-Qin Xu and Yuhei Inamori

ABSTRACT
As a new sludge reduction technology with a phosphorus removal mechanism, a vibration milling technology that uses iron balls have been applied to the wastewater treatment process. Three anaerobic–aerobic cyclic activated sludge processes: one without sludge disintegration; one disintegrated sludge by ozonation; and the other disintegrated sludge with the vibrating ball mill were compared. Ozonation achieved the best sludge reduction performance, but milling had the best phosphorus removal. This is because iron was mixed into the wastewater treatment tank due to abrasion of the iron balls, leading to settling of iron phosphates. Thus, the simple means of using iron balls as the medium in a vibrating ball mill can achieve both a sludge reduction of half and excellent phosphorus removal. Material balances in the processes were calculated and it was found that carbon components in disintegrated sludge were more resistant to biological treatment than nitrogen.

Key words | activated sludge, disintegration, excess sludge, phosphorus removal, vibration milling, wastewater treatment

INTRODUCTION
The activated sludge process is a common method of treating organic wastewater; however, as wastewater is processed, excess sludge is produced and problems such as rising costs of disposing this sludge and lack of land in which to bury the ash are attracting attention (Foley et al. 2010). Many lines of research into reducing excess sludge are currently being pursued, as grouped in Table 1 (Mitekura et al. 2002). In our new technology Tech. 4 listed in Table 1, produced sludge is converted to biodegradable components by disintegration processing and the disintegrated sludge is then circulated back to the wastewater treatment tank for further biodegradation. It has been reported that introduction of a sludge disintegrating technology increases the burden on the wastewater treatment tank and that water quality of the treated wastewater deteriorates due to disintegrated components of sludge that should have been discharged from the system as excess sludge (Sakai et al. 1997). In order to prevent deterioration in water quality of treated wastewater that is associated with sludge disintegration, introducing nitrogen removal (Dytczak et al. 2007) and phosphorus removal (Saktaywin et al. 2005) together with the sludge reduction technologies have been studied, whereas other studies have been conducted into combining sludge disintegration with advanced treatment technologies such as membranes and the like, which are grouped under Tech. 5 in Table 1 (Banu et al. 2009).

As a new sludge reduction technology based on disintegration that may be introduced into current wastewater treatment facilities, the vibrating ball mill has a strong
the introduction of contaminants. In a bead mill, which is in the same category of physical disintegrating technologies, ceramic balls are used for the grinding medium (Nawa 2002), but in our vibrating ball mill, iron balls are used. Our goal was to develop a sludge reduction technology with a phosphorus removal function by utilizing the iron that is supplied by abrasion of the iron balls as a secondary effect during milling and seeking the effect of iron phosphates precipitating in the wastewater treatment tank.

Operational conditions of vibration milling that are suitable for sludge grinding have been investigated previously (Sano et al. 2005). From our previous findings when applying a vibrating ball mill to a wastewater treatment process, it is important to ascertain wastewater treatment performance and the material balance of the overall wastewater treatment process. Previous research gives studies of comparison before and after introduction of a sludge reduction technology in Table 1 (Zhang et al. 2002). There are reports of comparative evaluations by simulation of the individual technologies of Table 1 (Mitekura et al. 2003), but no reports of practical comparisons by parallel testing of different sludge reduction technologies were found yet. Therefore, a comparative study of three systems was performed: an anaerobic–aerobic cyclic activated sludge process with no disintegration process; a process in which ozonation, which is a widely studied chemical disintegrating technology, was introduced into the anaerobic–aerobic cyclic activated sludge process; and a process in which vibration milling is employed in the anaerobic–aerobic cyclic activated sludge process. Our results clarified the sludge reduction performances and wastewater treatment performances, and the evaluations of each technologies were compared by calculating material balances of carbon, nitrogen, and phosphorus.

**METHODS**

**Wastewater treatment test (control)**

Figure 1 shows the general structure of the activated sludge test apparatus. The effective volume of the reactor was 24 L. Division plates were placed in the reactor to divide it into an anaerobic region (9.6 L) and an aerobic region (14.4 L). A basin with an effective volume of 7 L was used as a pre-effluent settling tank. Domestic wastewater that had been stored at 5 °C was supplied to the reactor by a supply pump. The supply flow rate (Qin) was controlled to 47 L d⁻¹ by a dispenser equipped with a surface level sensor. Accordingly, retention times in the anaerobic region and the aerobic region were 5.1 and 7.7 h, respectively. The reactor was equipped with a means for internal circulation and sludge circulation. As shown in Figure 1, an internal circulation pump connected an aerobic region with an anaerobic region and had a flow rate of 71 L d⁻¹ (1.5 Qin), and a sludge circulation pump connected the settling tank with an anaerobic region and had a flow rate of 24 L d⁻¹ (0.5 Qin). In order to keep the MLSS (mixed liquor suspended solids) homogeneous, low-speed stirrers were provided in anaerobic regions. For aerating and stirring aerobic regions, diffusers with flow rates of 2.3 L min⁻¹ were provided in the aeration regions. Excess sludge was discharged once a week to keep the MLSS in the reactor at 4,000 mg L⁻¹.

In the control system, the sludge reduction process shown in Figure 1 was not included. Therefore, all of the discharged sludge was discharged out of the system.

**Table 1 | Excess sludge reduction technologies grouped by method (Mitekura et al. 2002)**

<table>
<thead>
<tr>
<th>Fundamentals</th>
<th>Methods</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tech. 1 Decrease of sludge yield conversion ratio</td>
<td>Humus soil, field horsetail, porous carrier, acid, alkali</td>
</tr>
<tr>
<td>Tech. 2 Increase of self-oxidation coefficient</td>
<td>Oxidation ditch process, contact bed process, protozoa coexist</td>
</tr>
<tr>
<td>Tech. 3 High MLSS (mixed liquor suspended solids)</td>
<td>Membrane separation, saponin addition</td>
</tr>
<tr>
<td>Tech. 4 Solubilization and substrate production + biodegradation</td>
<td>Ozonation, mill, ultrasonic, hydrothermal reaction, digestion</td>
</tr>
</tbody>
</table>

**Figure 1 | Experimental activated sludge reactor for wastewater treatment.**
Ozonation (ozonating system)

The wastewater treatment test method was the same as the control system described above, except that the activated sludge reactor included an ozonation process. All discharged sludge was ozonated using a batch reactor. A cylindrical reactor with an effective volume of 2 L equipped with an antifoaming device at the top was used for ozone processing. See previous research from this laboratory for the ozonation conditions (Suzuki et al. 2006). The ozone concentration was set to 40 mg L\(^{-1}\) and the supply was regulated to 75 mg-consumed ozone-g-SS\(^{-1}\). The ozonated sludge was supplied to activated sludge reactor in the same manner as the domestic wastewater.

Vibrating ball mill (milling system)

The wastewater treatment test method was the same as the control system described above, except that the activated sludge reactor included a vibration milling process. Of the discharged sludge, 75% was milled using a batch milling for 15 min and the rest (25%) was discharged out of the system. For grinding conditions, see previous research (Sano et al. 2005). The rotation speed and amplitude of vibration were set to 750 rpm and 350 mm, respectively. Iron balls of 7.9 mm diameter were used for milling medium. Sample amounts and ball packing rates were set to optimum conditions obtained from the previous research. The ground sludge was supplied to activated sludge reactor in the same manner as the domestic wastewater.

Analysis

Water quality and sludge characteristics (BOD, TOC, TN [total nitrogen], TP [total phosphorus], SS, and VSS [volatile suspended solids]) were analyzed in accordance with the standard methods described in detail by the American Public Health Association (APHA/AWWA/WEF 2005). Soluble TOC, TN, and TP (DOC [dissolved organic carbon], D-TN [dissolved TN], and D-TP [dissolved TP]) were analyzed after filtering with a 0.45 μm filter (Millipore, MA). Ammonium (NH\(_4^+\)), orthophosphate (PO\(_4^{3-}\)), and nitrate and nitrate (NO\(_2^-\), NO\(_3^-\)) were analyzed using a TrAacs 2000 analyzer (Bran+Luebbe K.K., Japan). Iron concentration was subjected to acid decomposition following the method of Hou et al. (2006) and was then analyzed with ICP-AES (ICAP-750, Nippon Jarrell-Ash, Japan).

RESULTS AND DISCUSSION

Sludge reduction effect and characteristics of disintegrated sludge

The characteristics of the sludges disintegrated by ozonation and by vibration milling were compared. Table 2 shows changes in the characteristics of the sludges according to the two kinds of processing. The processes were operated stably for half a year and the values in the table are average values (n = 8) over the last two months. Each value represents a ratio between before and after processing.

Because the SS components of the sludge were disintegrated by the two kinds of processing, SS\(_{\text{after}}\)/SS\(_{\text{before}}\) was low in both cases. Total concentrations of each component (TOC, TN, and TP) were not changed by the two kinds of processing. Therefore, it can be suggested that sludge components were not broken down into gases under the test conditions. DOC\(_{\text{after}}\)/DOC\(_{\text{before}}\) and D-TN\(_{\text{after}}\)/D-TN\(_{\text{before}}\) were raised by factors of 12.1 and 2.4, respectively, by the ozonation, and were raised by factors of 6.2 and 2.3 by the vibrating ball mill. It can be suggested that this is because the ozonation and milling ruptured cell membranes releasing solutes in the cells, which concurs with previous findings (Chu et al. 2009; Wang et al. 2010). Ozonation, with the lower SS\(_{\text{after}}\)/SS\(_{\text{before}}\) ratio, gave rising rises in DOC and D-TN compared with milling. D-TP\(_{\text{after}}\)/D-TP\(_{\text{before}}\) rose by 2.7 with ozonation but fell by 0.9 with milling. Only D-TP\(_{\text{after}}\)/D-TP\(_{\text{before}}\) with milling showed a different behavior from the other soluble components.

Table 3 shows a comparison of excess sludge amounts X\(_i\), and VSS/SS ratio obtained from the wastewater treatment tests.

The sludge production amount in the control system was 5.5 g-SS d\(^{-1}\). The excess sludge amount in the ozonation system was 0.2 g-SS d\(^{-1}\), which is very low, reaching a sludge reduction of 96%. The excess sludge amount in the
milling system was 2.6 g-SS d\(^{-1}\), a sludge reduction of 52%. In previous research, bead milling has provided a sludge reduction of 80% (Nawa 2002). It can be suggested that the rate with the vibration milling of this study is not as high as this because precipitated iron phosphates are included in the sludge.

VSS/SS was 0.82 in the control system, 0.79 in the ozonation system, and 0.72 in the milling system. Two sludge reduction systems gave lower values than the control system. This is because inorganic matter discharged out of the system as excess sludge accumulates in the wastewater treatment tank for sludge reduction system as reported in previous findings (Sakai et al. 1997; Hirooka et al. 2006). Although the ozonation system, which has large circulation amounts of disintegrated sludge, would be expected to have the lowest VSS/SS, because iron abraded by the milling is contained in the sludge, at around 0.09 g-Fe g-SS\(^{-1}\), VSS/SS was lower in the milling system.

### Table 3: Comparison of excess sludge production and VSS/SS

<table>
<thead>
<tr>
<th></th>
<th>Control</th>
<th>Ozone</th>
<th>Mill</th>
</tr>
</thead>
<tbody>
<tr>
<td>(X_f)</td>
<td>[g-SS day(^{-1})]</td>
<td>5.5</td>
<td>0.2</td>
</tr>
<tr>
<td>VSS/SS</td>
<td>[g-VSS g-SS(^{-1})]</td>
<td>0.82</td>
<td>0.79</td>
</tr>
</tbody>
</table>

Comparison of water quality of treated wastewater

It is clear that the circulation of disintegrated sludge associated with introduction of a sludge disintegrating technology has an effect on wastewater treatment performance. Therefore, the water qualities of the treated wastewaters were compared. The water qualities of influent wastewater and treated water effluent for each system are shown in Table 4. There were no great differences in the treated water BOD and SS for different systems. The treated water TOC and DOC were higher in the two sludge reduction systems than in the control system as reported by Yasui (Yasui et al. 1996). The ozonation system, with large circulation amounts of disintegrated sludge, was significantly worse than the milling system. DOC and TOC of treated water showed almost the same values for each system. From these results, the organic matter in treated wastewater would almost be composed of dissolved organic matter that is resistant to biodegradation. It can be suggested that the circulation of disintegrated sludge with the sludge reduction systems led to organic matter that is resistant to biodegradation, such as cell walls, being discharged without being decomposed.

### Table 4: Performances of wastewater treatment for different sludge reduction methods

<table>
<thead>
<tr>
<th></th>
<th>Influent</th>
<th>Effluent</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Control</td>
<td>Ozone</td>
</tr>
<tr>
<td>BOD</td>
<td>[mg L(^{-1})]</td>
<td>159</td>
</tr>
<tr>
<td>TOC</td>
<td>[mg L(^{-1})]</td>
<td>154</td>
</tr>
<tr>
<td>DOC</td>
<td>[mg L(^{-1})]</td>
<td>90</td>
</tr>
<tr>
<td>TN</td>
<td>[mg L(^{-1})]</td>
<td>44</td>
</tr>
<tr>
<td>NH(_4)-N</td>
<td>[mg L(^{-1})]</td>
<td>30</td>
</tr>
<tr>
<td>NO(_2)-N</td>
<td>[mg L(^{-1})]</td>
<td>ND</td>
</tr>
<tr>
<td>TP</td>
<td>[mg L(^{-1})]</td>
<td>4.5</td>
</tr>
<tr>
<td>D-TP</td>
<td>[mg L(^{-1})]</td>
<td>2.3</td>
</tr>
<tr>
<td>SS</td>
<td>[mg L(^{-1})]</td>
<td>125</td>
</tr>
</tbody>
</table>

NH\(_4\)-N of the treated water was the same in all the systems, with the nitrification being excellent in each system. TN and NO\(_2\)-N of the treated water showed the same values for all systems.

TP of the treated water was higher in the ozonation system (3.2 mg L\(^{-1}\)) than in the control system (2.7 mg L\(^{-1}\)) but was much lower in the milling system (1.2 mg L\(^{-1}\)). With the ozonation system, similarly to previous findings, phosphorus removal performance fell. It can be suggested that the reason excellent phosphorus removal performance was provided by the milling system is because iron was mixed into the wastewater treatment tank as a secondary effect, by abrasion of the iron balls during the milling. It was suggested that mixing iron caused flocculation of iron phosphates in the wastewater treatment tank. This phenomenon corresponded with phosphorus removal using iron electrolysis (Stanford et al. 2010) and packed bed of iron particles (Tashiro & Sakakibara 2005). Thus, by the simple means of using iron balls as the medium in the vibrating ball mill, both sludge reduction of half and excellent phosphorus removal were successfully provided. Furthermore phosphorus removal was stabilized and was unaffected by Fe mixing in this long-term operation.

### Material balances of carbon, nitrogen, and phosphorus

The wastewater treatment tests were operated continuously for a year to estimate material balances at steady state. The discharged sludge amounts and VSS/SS of sludge had stabilized after half a year and over the last two months, respectively. From these results over the last two months, material balances of carbon, nitrogen, and phosphorus were calculated in the control, ozonation and milling systems, respectively. Material balances of the systems are
shown in Figures 2–4. The systems were divided into:
(1) wastewater influent, (2) biological reaction, (3) treated water effluent, (4) sludge discharge, and (5) gases mineralized (removed) by the biological reaction, and respective values using the TOC, TN and TP values shown in Tables 2–4 were calculated. The amounts of mineralized (removed) carbon and nitrogen and the amount of phosphorus accumulated in the biological reaction were obtained by subtracting the treated water effluent and sludge discharge values from the wastewater influent values. Where a sludge disintegration technology was included, the results in Tables 2 and 3 were used to find the changes in soluble components caused by ozonation and the vibrating ball mill, which are shown in parentheses. See previous reports for the calculation methods (Tsuno et al. 2007; Arakawa & Tanaka 2008).

The mineralized (gasiﬁed) amounts of carbon and nitrogen were higher in both of the sludge reduction systems than in the control system. Thus, it can be seen that introduction of a sludge reduction technology promoted mineralization (gases). The amount of mineralized (gasiﬁed) carbon is higher with the milling system, and the amount of mineralized (gasiﬁed) nitrogen is higher with the ozonation system. It can be suggested that this result relates to changes in the C/N ratio associated with introduction of the sludge disintegrating technologies.

The differences in amounts of carbon discharged as treated water between sludge reduction systems and control system were +0.23 g d⁻¹ for the ozonation system and +0.05 g d⁻¹ for the milling system, respectively. The increases in carbon in the treated water effluent relative to sludge disintegrating amounts (differences between influent and effluent amounts of soluble components at the ozone processing and the mill processing) were 68 and 36% in the ozonation system and the milling system. Thus, it is clear that, of amounts of carbon in the disintegrated sludge, 32% was decomposed by biological reaction in the ozonation system and 64% was decomposed by biological reaction in the milling system.

The differences in amounts of nitrogen discharged as treated water between sludge reduction systems and control system were +0.06 g d⁻¹ for the ozonation system and +0.04 g d⁻¹ for the milling system. The increases in nitrogen in the treated water effluent were 8 and 11% in the ozonation system and the milling system. Thus, it is clear that, of amounts of nitrogen in the disintegrated sludge, most (92 and 89%) was decomposed by biological reaction in the ozonation system and 64% was decomposed by biological reaction in the milling system.

The differences in amounts of carbon discharged as treated water between sludge reduction systems and control system were +0.23 g d⁻¹ for the ozonation system and +0.05 g d⁻¹ for the milling system, respectively. The increases in carbon in the treated water effluent relative to sludge disintegrating amounts (differences between influent and effluent amounts of soluble components at the ozone processing and the mill processing) were 68 and 36% in the ozonation system and the milling system. Thus, it is clear that, of amounts of carbon in the disintegrated sludge, 32% was decomposed by biological reaction in the ozonation system and 64% was decomposed by biological reaction in the milling system.

The differences in amounts of nitrogen discharged as treated water between sludge reduction systems and control system were +0.06 g d⁻¹ for the ozonation system and +0.04 g d⁻¹ for the milling system. The increases in nitrogen in the treated water effluent were 8 and 11% in the ozonation system and the milling system. Thus, it is clear that, of amounts of nitrogen in the disintegrated sludge, most (92 and 89%) was decomposed by biological reaction.

These two effects show that, when a sludge disintegrating technology is included, carbon components contained in the disintegrated sludge, such as cellulose which is a component of cell walls, are more resistant to biodegradation than nitrogen components such as proteins contained in the disintegrated sludge.
The differences in amounts of phosphorus discharged as treated water between sludge reduction systems and control system were +0.02 g d\(^{-1}\) for the ozonation system and -0.07 g d\(^{-1}\) for the milling system.

**CONCLUSIONS**

As a new sludge reduction technology based on disintegration that can be introduced into existing wastewater treatment facilities, we developed a vibrating ball mill that uses iron balls. The technology was introduced into a wastewater treatment process, and the sludge reduction performance and wastewater treatment performance were evaluated. A comparative study of three systems: an anaerobic–aerobic cyclic activated sludge process as a control; an anaerobic–aerobic cyclic activated sludge process including a process for disintegrated sludge by ozonation; and an anaerobic–aerobic cyclic activated sludge process including a process for disintegrated sludge by the vibrating ball mill were conducted.

The ozonation system achieved a sludge reduction of 96% compared with a reduction of 52% in the milling system. Comparing water quality of treated wastewater revealed that with a sludge reduction system, dissolved organic matter resistant to biodegradation was discharged as wastewater without being decomposed and DOC of the treated water was worse. No impediment to nitrification and denitrification performance reaction were apparent in both of the sludge reduction systems. TP of the treated water was higher in the ozonation system than in the control system, and was lower in the milling system. It can be suggested that the reason the milling system provided a better phosphorus removal performance was that, as a secondary effect during the milling, iron was mixed into the wastewater treatment tank due to abrasion of the iron balls, leading to precipitation of iron phosphates. Thus, by the simple means of using iron balls as the medium in the vibrating ball mill, both sludge reduction of half and excellent phosphorus removal performance could be obtained. From these results, material balances of carbon, nitrogen, and phosphorus in control, ozonation, and milling systems were calculated. The results showed that amounts of mineralized (removed) carbon and nitrogen were higher in both of the sludge disintegrating systems than in the control system, and mineralization (removal) was promoted by introduction of a sludge disintegrating technology. In addition, it was clear that when a sludge disintegrating technology was included, carbon components contained in disintegrated sludge were more resistant to biodegradation than nitrogen components.

**REFERENCES**


First received 7 July 2011; accepted in revised form 26 August 2011