Decomposition of excess sludge in a membrane bioreactor using a turbulent jet flow ozone contactor

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1. Introduction

The membrane bioreactor (MBR) process is one of the most economical and promising technologies for domestic and industrial wastewater treatment [1]. In conventional wastewater treatment plants, the handling and disposal of sludge can account for 25–65% of the total operation costs [2]. Although the MBR process generates lesser amounts of biomass (or excess sludge) because of its high solids retention time (SRT), the excess sludge produced in the MBR still creates a concurrent sludge management problem [3]. Therefore, it would be desirable to develop an efficient means of reducing the levels of excess sludge in the MBR.

One solution to the problem of excess sludge would be installing a pretreatment system prior to the MBR. Among the various reported pretreatment methods – including thermal, ultrasonic, mechanical, alkaline, and oxidative technologies [4–6] – ozonation appears to be one of the most efficient because of its high solids retention time (SRT), the excess sludge produced in the MBR still creates a concurrent sludge management problem [3]. Therefore, it would be desirable to develop an efficient means of reducing the levels of excess sludge in the MBR.

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1.1. Sludge decomposition process

Fig. 1 displays the process of sludge decomposition by the TJC. First, the raw excess sludge is broken down into smaller pieces that may be further pulverized into primary particles (e.g., small aggregates of microbial cells), in a process known as “sludge disintegration” [14]. Second, the disintegrated primary particles...
are further destroyed, releasing soluble organic matter (e.g., cytoplasmic materials), in the so-called “solubilization” step. Third, the solubilized organics are further oxidized into minerals \((CO_2, H_2O)\) in the “mineralization” process. Thus, the sludge decomposition sequence comprises disintegration, solubilization, and mineralization \([9, 15]\). In this study, “sludge reduction” refers to the removal of mixed liquor suspended solids (MLSSs) through solubilization and/or mineralization of the raw sludge. We determined the degree of sludge reduction from the difference in MLSS concentrations of the sludge before and after treatment with the TJC. Likewise, we estimated the extent of mineralization from the difference in the total CODs (TCOD) before and after treatment of the raw sludge.

2. Materials and methods

2.1. TJC system

The laboratory-scale TJC (OXEN Tech Co., Ltd., Seoul, Korea) comprised five major compartments (Fig. 2): (i) an ozone and liquid contactor (working volume: 30 L) equipped with a baffle plate, (ii) a venturi injector, (iii) a circulation pump, (iv) oxygen and ozone generators, and (v) an off-gas ozone destructor. Oxygen was converted into ozone through an ozone generator (VMUS-08E, AZCO Industries, Ltd., Langley, Canada). The excess sludge withdrawn from the TJC-MBR was fed to the TJC for sludge reduction in batch mode at an ozone concentration of 68 mg-O3 L\(^{-1}\). The sludge was recirculated, passing through the orifice of the venturi injector (diameter: 5 mm) at a flow rate of \(30 \pm 1\) L min\(^{-1}\) to generate hydrodynamic cavitation. As long as the mixed liquor was being circulated, the ozone gas was suctioned automatically and continuously into the contactor via the venturi injector as a result of the development of negative pressure. During TJC operation, samples were removed periodically from the TJC to monitor the degree of sludge decomposition, which was subdivided into degrees of sludge disintegration, solubilization, and mineralization. The maximum ozone contact time for excess sludge treatment was 60 min. At the end of this period of time, the ozone transfer efficiency was ca. 96% at an ozone concentration of 68 mg-O3 L\(^{-1}\) in the gas phase. The converted specific ozone (SO) dosage for TJC operation was 1.28 mg-O3/g-MLSS min.

Hydrodynamic cavitation is the sequential generation, growth, and collapse of cavitation bubbles, releasing high-energy waves at the surface of the bubbles for a very short period of time. The cavitation bubbles implode within milliseconds under the high pressure surrounding them, releasing tremendous amounts of energy in the form of shock waves \([16]\). Highly oxidizing radicals (e.g., \(HO^\cdot\)) can be generated from these waves, analogous to those formed through advanced oxidation processes (AOPs) \([17–19]\).

A dimensionless cavitation number is used to characterize the process; under ideal conditions, hydrodynamic cavitation is generated when its value is less than 1 \([16]\). In the TJC system, hydrodynamic cavitation was achieved by forcing the excess sludge through an annular opening that combined a narrow entrance orifice with a much larger exit orifice (Fig. 2); the cavitation number for the TJC system was calculated to be within the range 0.24–0.25. Moreover, the baffle plate at the lower part of the contactor could smash the fluid, causing a certain portion of it to flow back, thereby reinforcing the turbulent flow. As a result, the TJC system maximized both the disintegration of the sludge particles and the ozone transfer efficiency.

2.2. Sample collection and determination

Samples were collected periodically from the TJC-MBR to monitor the degree of sludge decomposition, which was subdivided into degrees of sludge disintegration, solubilization, and mineralization. The maximum ozone contact time for excess sludge treatment was 60 min. At the end of this period of time, the ozone transfer efficiency was ca. 96% at an ozone concentration of 68 mg-O3 L\(^{-1}\) in the gas phase. The converted specific ozone (SO) dosage for TJC operation was 1.28 mg-O3/g-MLSS min.

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**Fig. 2.** Schematic representation of the Control-MBR and TJC-MBR systems.
Table 1
Composition of synthetic wastewater.

<table>
<thead>
<tr>
<th>Component</th>
<th>Concentration (mg L⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glucose</td>
<td>160</td>
</tr>
<tr>
<td>Bacto peptone</td>
<td>120</td>
</tr>
<tr>
<td>Yeast extract</td>
<td>16</td>
</tr>
<tr>
<td>(NH₄)₂SO₄</td>
<td>104</td>
</tr>
<tr>
<td>KH₂PO₄</td>
<td>33</td>
</tr>
<tr>
<td>MgSO₄·7H₂O</td>
<td>32</td>
</tr>
<tr>
<td>MnSO₄·4H₂O</td>
<td>2.8</td>
</tr>
<tr>
<td>FeCl₃·6H₂O</td>
<td>0.1</td>
</tr>
<tr>
<td>CaCl₂·2H₂O</td>
<td>3.2</td>
</tr>
<tr>
<td>NaHCO₃</td>
<td>220–500</td>
</tr>
<tr>
<td>MnSO₄·7H₂O</td>
<td>32</td>
</tr>
</tbody>
</table>

2.2. TJC-MBR system

Fig. 2 presents a schematic representation of the Control- and TJC-MBRs. Two MBRs, each having a working volume of 50 L, were operated in parallel at room temperature 25 (±2) °C. The same synthetic wastewater (see Table 1 for its composition) was fed into both MBRs, except that, after being disintegrated by the TJC system, all of the excess sludge was recycled only to the TJC-MBR.

A membrane module (area: 0.38 m² module⁻¹) was mounted vertically in each MBR (Fig. 2). Each module was prepared from polyvinylidene fluoride (PVDF) hollow fiber membranes (Ge-Zenon, US) having a pore size of 0.04 µm. A coarse bubble diffuser was placed at the bottom of each MBR to maintain a dissolved oxygen concentration of 2.5–3.0 mg L⁻¹. The filtration was stopped when the transmembrane pressure (TMP) reached 30 kPa. All of the membrane modules in each reactor were operated continuously at a constant flux of 13 L m⁻² h⁻¹. A portion of the permeate was recycled to the reactor to maintain a constant working volume of 50 L so that the hydraulic retention time (HRT) was maintained at 10 h in each MBR.

Once a day, 2.5 L of the mixed liquor in both MBRs was removed to maintain a sludge retention time (SRT) of 20 days. The excess sludge from the TJC-MBR was collected and preserved at 4 °C until the total accumulated volume reached 30 L [20]. The collected sludge was disintegrated in the TJC system in a batch mode and then 2.5 L of this treated sludge was recycled to the TJC-MBR once a day; in contrast, the excess sludge from the Control-MBR was discarded.

2.3. Analytical methods

The MLSS and the mixed liquor volatile suspended solids (MLVSSs) were measured using standard methods [21]. The total COD (TCOD) and soluble COD (SCOD) of the mixed liquor were measured using a DR4000 spectrophotometer (Hach Co., Denver, CO, USA) and corresponding reagent kits. The ozone concentration in the gas phase was monitored using an ozone gas analyzer (H1-S, IN USA Corp., Norwood, MA, USA). The particle size distribution of the microbial flocs (i.e., sludge particles) in the mixed liquor was monitored using a particle size analyzer (MasterSizer/E, Malvern Instruments, Ltd., Malvern, UK). The smaller sludge particles (<8 µm) in the decomposed sludge were determined using a dynamic light scattering method (ELS-Z, Otsuka, Japan) after filtration through an 8-µm MF membrane (Mixed cellulose esters, Hydrophilic, Millipore, Billerica, MA, USA).

3. Results and discussion

3.1. Effect of TJC pretreatment on mixed liquor characteristics

Table 2 summarizes the characteristics of the mixed liquors in the raw excess sludge and in the sludge decomposed using the TJC; the latter is further subdivided into systems with ozonation (CAV-OZO sludge) and without ozonation (CAV sludge). The values of pH of the mixed liquors in the raw excess sludge and the CAV sludge were 7.0 and 7.1, respectively. Notably, however, the pH of the mixed liquor in the CAV-OZO sludge dropped to 6.0 after ozonation with the TJC, confirming that mineralization, producing CO₂, had occurred. The SCOD increased from 78 mg L⁻¹ for the raw excess sludge to 200 and 2300 mg L⁻¹ for the CAV sludge and the CAV-OZO sludge, respectively. Although the hydrodynamic cavitation increased the SCOD to a certain degree, cavitation combined with ozonation greatly enhanced the sludge solubilization. Simultaneously, the TCOD decreased by 64% from 8700 to 3100 mg L⁻¹ after 60 min of TJC operation, indicating that mineralization of the sludge that had been subjected only to cavitation was negligible (<10%), whereas it was considerable (>60%) for the system that had experienced both cavitation and ozonation. The MLSS decreased by only 8% after cavitation, whereas it decreased by ca. 60% after both cavitation and ozonation; in contrast, the MLVSS decreased by ca. 18 and 77%, respectively, indicating that the combination of cavitation and ozonation decomposed organic solids more readily than inorganic ones.

Table 2
Characteristics of the raw excess sludge and the sludge decomposed using the TJC.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Raw excess sludge</th>
<th>Decomposed sludge (contact time: 60 min)</th>
<th>CAV sludge (cavitation only)</th>
<th>CAV-OZO sludge (cavitation + ozonation)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.0 (±0.3)</td>
<td>7.1 (±0.5)</td>
<td>7.1 (±0.5)</td>
<td>6.0 (±0.4)</td>
</tr>
<tr>
<td>SCOD (mg L⁻¹)</td>
<td>78 (±8)</td>
<td>199 (±11)</td>
<td>2330 (±91)</td>
<td>3120 (±216)</td>
</tr>
<tr>
<td>TCOD (mg L⁻¹)</td>
<td>8720 (±226)</td>
<td>8020 (±340)</td>
<td>3050 (±345)</td>
<td>3050 (±345)</td>
</tr>
<tr>
<td>MLSS (mg L⁻¹)</td>
<td>7625 (±134)</td>
<td>6990 (±122)</td>
<td>3050 (±345)</td>
<td>3050 (±345)</td>
</tr>
<tr>
<td>MLVSS (mg L⁻¹)</td>
<td>6213 (±142)</td>
<td>5107 (±75)</td>
<td>1364 (±67)</td>
<td>1364 (±67)</td>
</tr>
</tbody>
</table>

a Specific ozone dosage (SD) = 1.28 mg-O₃/g-MLSS min.
3.2. Sludge reduction by the TJC

Fig. 3 displays the extent of sludge reduction by the TJC system as a function of contact time. When only hydrodynamic cavitation was applied (i.e., CAV-TJC), the percentage sludge reduction was maintained at less than 8% for contact times up to 60 min; i.e., sludge decomposition through physical treatment alone, e.g., hydrodynamic cavitation, was ineffective. When both cavitation and ozonation (i.e., CAV/OZO-TJC) were applied, however, the percentage sludge reduction increased rapidly to ca. 20% within 3 min and reached more than 60% after 60 min, suggesting that sludge decomposition occurred extensively during the early stages of TJC operation. These results suggest that cavitation and ozonation had a synergic effect on the degree of sludge decomposition in the TJC system.

To compare the efficiency of our sludge reduction method using the TJC with those performed previously using other ozone treatment processes [22], we plotted them as functions of the ozone dosage (Fig. 4a) and the specific energy (Fig. 4b). In Fig. 4a, the ozone dosage is given on weight basis (mg of ozone per g of MLSS). The TJC system provided 10–20% higher degrees of sludge reduction relative to the other reported ozone treatment systems. At an ozone dosage of 80 mg-O3/g-MLSS, sludge reduction with the TJC reached 60%, whereas it was 35–45% when using the other systems. We attribute the higher degrees of sludge reduction, when using the TJC, to the effective ozone transfer efficiency and the great extent of sludge disintegration as a result of hydrodynamic cavitation.

Although the generation of hydrodynamic cavitation is one advantage of the TJC system, it requires an additional power source for the recirculation pump, one that is not necessary in the other ozone treatment systems. Therefore, it was important for us to compare the degrees of sludge reduction of the various ozone treatment systems in terms of their specific energy inputs. The specific energy ($E_{spec}$) provides information on the necessary energy input to achieve a certain degree of sludge reduction. It is defined as the energy input per mass of ozonated sludge, and is expressed using the following equation [22,23]:

$$E_{spec} = \frac{P \cdot t}{V \cdot MLSS}$$  \hspace{1cm} (1)

where $P$ is the sum of energies consumed (W) by the four different power sources (oxygen and ozone generators, ozone gas analyzer, and circulation pump), $t$ is the time for ozone reaction (s), $V$ is the sludge volume (L), and $MLSS$ is the mixed liquor suspended solid concentration (g L$^{-1}$).

All the energies consumed for the TJC were summed and compared with those for the other ozonation systems reported in the literature (Fig. 4b). Even when considering the additional power source (i.e., circulation pump), the TJC provided 5–25% higher degrees of sludge reduction, depending on the given specific energy. In particular, the greater the specific energy input, the
higher the difference in sludge reduction between the TJC and the other systems.

3.3. Solubilization and/or mineralization of the sludge during TJC operation

The ultimate goal of the TJC system for the pretreatment of excess sludge is to establish a zero-discharge MBR system by completely recycling the decomposed sludge to the MBR. The SCOD in the treated sludge contains a certain fraction of biodegradable organic matter, which contributes to the ratio of food to microorganisms (F/M). Moreover, the SCOD may affect the membrane filterability in an MBR [24]. Consequently, the SCOD of the decomposed sludge would certainly be a key factor for the establishment of a zero-discharge MBR system. In addition, the change in TCOD of the sludge before and after treatment with the TJC is another important parameter because it indicates the mineralization capability of the system.

In this context, Fig. 5a and b displays the levels of SCOD and TCOD, respectively, monitored during operation of the TJC. Within 3 min, the SCOD increased from 78 mg L\(^{-1}\) in the raw excess sludge to 150 mg L\(^{-1}\) in the sludge treated with cavitation only (CAV-TJC), indicating that the CAV-TJC system had limited solubilization capability for particulate organic matter (POM). In contrast, the SCOD increased drastically in the sludge treated with both cavitation and ozonation (CAV/OZO-TJC), from 78 mg L\(^{-1}\) in the raw excess sludge to 1290 mg L\(^{-1}\) within 3 min; it leveled off at ca. 2500 mg L\(^{-1}\) after 25 min, implying that the CAV/OZO-TJC process led to very rapid and effective solubilization of the POM in the sludge.

On the other hand, little change in the TCOD occurred in the CAV-TJC system (Fig. 5b), suggesting that mineralization of the dissolved organic matter (DOM) was negligible under these conditions. For the CAV/OZO-TJC system, however, the TCOD decreased slightly, from 8700 to 8400 mg L\(^{-1}\), within the first 25 min, but decreased considerably thereafter, to 3100 mg L\(^{-1}\), after a contact time of 60 min. The starting point (ca. 25 min of operation time) of the substantial decrease in the TCOD (Fig. 5b) coincides well with the leveling-off point (ca. 25 min of operation time) of the SCOD (Fig. 5a). In other words, the dissolved organic matter (DOM) in the mixed liquor underwent mineralization after 25 min, leading not only to leveling-off of the SCOD (rather than its continuous increase) but also to reduction of the TCOD.

3.4. Disintegration of sludge particles during TJC operation

To investigate the ability of the TJC to disintegrate and/or solubilize sludge particles (e.g., microbial flocs), we monitored the mean particle sizes of sludge particles with respect to the contact time under the CAV-TJC and CAV/OZO-TJC conditions (Fig. 6). The
mean particle size of the raw excess sludge taken from the MBR was ca. 240 μm, but it dropped rapidly (<1 min) to 105 and 68 μm in the CAV-TJC and CAV/OZO-TJC systems, respectively. Although the CAV/OZO-TJC condition disintegrated the sludge particles faster than CAV-TJC did during the initial stages, in both cases the mean particle size dropped to ca. 38 μm after 15 min and leveled off thereafter. Thus, hydrodynamic cavitation played a major role in the disintegration of sludge particles.

To acquire more information on the change in the particle size distribution during TJC operation, we classified the sizes of the particles into three levels, based on the approach used in a previous report [25]: microparticles (d1; <1.2 μm), intermediate particles (d2; 1.2–50 μm), and macroparticles (d3; >50 μm). Particles d1 and d3 include primary particles and sludge flocs, respectively; d2 covers secondary particles that link primary particles with extra polymeric substances (EPS) [25]. Fig. 7 displays their volumetric fractions plotted with respect to the contact time.

Fig. 7 reveals that particles larger than 50 μm (d3) underwent the most intense disintegration within the initial 3 min. After the first 1 min of contact time, the volume percentage of d3 dropped from 90 to 73% under CAV-TJC conditions (Fig. 7a) and to 62% under CAV/OZO-TJC conditions (Fig. 7b). At the same time, the volume percentage of the intermediate particles (d2) increased rapidly from 9 to 25% under the CAV-TJC conditions (Fig. 7a) and to 38% under the CAV/OZO-TJC conditions (Fig. 7b). Accordingly, the fractional differences between d2 and d3 were 48 and 24%, respectively. In other words, the fractional difference between d2 and d3 in the CAV-TJC system without ozonation was double that in the CAV/OZO-TJC system with extra polymeric substances (EPS) [25]. Fig. 7 displays their volumetric fractions plotted with respect to the contact time.

On the other hand, the fractional volume percentage change for the microparticles (d1) was barely noticeable in either case (Fig. 7), primarily because they were too small to observe any change. Therefore, we further divided the sizes of the microparticles into two groups: d1, (<0.45 μm) and d1, (0.45–1.2 μm).

Fig. 8 plots the relative fractional changes of these two types of microparticles. For the CAV-TJC system (Fig. 8a), the fraction of the smaller microparticles (d1) increased from <1 to 34% within 3 min and then increased gradually to 50% after 60 min. For the CAV/OZO-TJC system (Fig. 8b), however, the fraction of d1 jumped to 90% within 10 min and leveled off thereafter. Thus, the fraction d1 (90%) in the CAV/OZO-TJC system was much greater than that (50%) in the CAV-TJC system, consistent with the results in Fig. 7. It also suggests that the cavitation and ozonation reactions have a synergetic effect on sludge disintegration and/or solubilization in the CAV/OZO-TJC system. This conclusion agrees with the much higher release of SCOD in the CAV/OZO-TJC system than that in the CAV-TJC system, as indicated in Fig. 5a.

3.5. Effect of disintegrated sludge recycling on TMP

The rate of TMP build-up is an important factor when evaluating membrane filterability in a submerged MBR because it is directly related to the extent of membrane fouling [28–30]. Fig. 9 presents the variations in TMP plotted with respect to the operating time in both the Control- and TJC-MBRs. In the TJC-MBR, the time required to reach a TMP of 30 kPa (14 days) was nearly double that (8 days) required in the Control-MBR (cf. points 1 and 2 in Fig. 9). In other words, the recycling of disintegrated sludge to the MBR after treatment with the TJC reduced the rate of membrane fouling substantially.

4. Conclusions

In this study, we investigated the decomposition of excess sludge generated in an MBR incorporating a TJC, which performed
the combined functions of mechanical disintegration (hydrodynamic cavitation) and chemical oxidation (ozonation). The following conclusions are drawn:

(1) The TJC system provided 10–20% higher degrees of sludge reduction at a given ozone dose relative to those of other reported ozone treatment systems. Furthermore, even with its requirement of an additional power source (e.g., circulation pump), the TJC resulted in 5–25% higher degrees of sludge reductions at a given specific energy. The greater the specific energy input, the greater the difference in the degrees of sludge reduction between those of the TJC and other systems.

(2) Hydrodynamic cavitation played a major role in the disintegration of the sludge particles because it gave rise to a higher ozone mass transfer efficiency, which in turn promoted the disintegration of the sludge particles in the TJC.

(3) Recycling of the disintegrated sludge to the MBR after treatment with the TJC reduced the rate of membrane fouling substantially.

Acknowledgment

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References