A novel approach to treat combined domestic wastewater and excess sludge in MBR

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Abstract: Domestic wastewater was treated by combined anaerobic biofilm-aerobic membrane bioreactor (MBR) process, and part biomass in MBR was withdrawn to treat with ozone, then the ozonated sludge was returned to anaerobic inlet. In aerobic MBR, MLSS and DO were controlled at 3000—3500 mg/L and 0.8 mg/L respectively. Comparing the experimental results of two stages, it was noticed that ozonation did not affect the removal efficiency for organics but had a significant influence on the removals of NH$_3$-N and TN. During the ozonation period of two months, no excess sludge was wasted, and a zero sludge yield was obtained.

Keywords: combined anaerobic biofilm-aerobic membrane (MBR); domestic wastewater; ozonation; excess sludge; sludge yield

Introduction

In a conventional wastewater treatment plant, large amounts of biosolids are formed and the separation, dewatering, treatment and disposal of this sludge represents major investment and operating costs. With the rising costs and restrictions on sludge disposal, the minimization of sludge yield has become of increasing importance (Chen, 2000). Therefore, it becomes interesting to modify conventional wastewater treatment process in order to reduce biosolids production.

In most cases, anaerobic process has always been used for the disposal of excess sludge in wastewater treatment plant. In addition, it has also been applied as the pretreatment of highly concentrated industrial wastewater. At present, the research on treating lowly concentrated domestic sewage with combined anaerobic and aerobic technology has also been conducted (Castillo, 1997). Compared with conventional aerobic technology, anaerobic process has two distinct advantages of low production of excess sludge and low energy demands.

In recent years, MBR has been proposed as an alternative for conventional activated sludge process. The advantage of MBR is mainly due to the fact that it can maintain high MLVSS in the reactor. It has gained increasing use in wastewater treatment because of its several advantages.

(1) The retention time of the biomass can be controlled as long as desired, which will create favorable conditions for normal growth of some species of bacteria with low growth rates, such as nitriﬁers. (2) Possible implementation of simultaneous nitrification and denitriﬁcation because of its highly concentrated MLVSS, which make it easy to form aerobic zone and anoxic zone in the same reactor. (3) Better and more reliable efﬂuent quality compared to that of conventional process and no need for post-treatment (Silva, 1998). (4) Easy automatic control and compactness of the whole system.

The aim of this research was to minimize the production of excess sludge. To achieve this goal, combined anaerobic biofilm with aerobic MBR process was selected to treat domestic wastewater, and part biomass in MBR would be treated with ozone, then the ozonated sludge was returned to the anaerobic inlet and mixed with influent for further biological treatment. This treatment process configuration had been proposed on the basis of following reasons: (1) Anaerobic process can effectively remove organics, at the same time, it can reduce the production of excess sludge as well as the energy consumption. Then the ozonated excess sludge was returned into the anaerobic tank, which can play an important role in degradation and stabilization of ozonated excess sludge. (2) Aerobic MBR can maintain a highly concentrated sludge, which is easy to implement simultaneous nitrification and denitriﬁcation in the same
reactor under low DO condition. (3) It was demonstrated that the net biomass growth could be reduced under cryptic conditions (Hamer, 1985; Mason, 1987), which is to say, when the organic carbon in microorganism is used as substrate of metabolism process, sludge production will be reduced. Based on these hypotheses, it was assumed that ozone could be used as a cell lysis agent, because ozone is a strong oxidation agent, it can promote the biodegradation of the cell, then the treated excess sludge was returned to the biological treatment unit, which will minimize the net excess sludge production.

1 Materials and methods

1.1 Experimental equipment

A hollow fiber polysulfone ultrafiltration (UF) membrane module with a total surface area of 1.5 m², pore size of 0.05 μm and a length of 0.35 m was used as the test membrane. The volumes of the anaerobic and aerobic reactor were 8.2 L and 16.3 L respectively. The temperature of both reactors was maintained at 20°C with a thermostat. A water float cock was used to control water level of the reactor to keep the balance of influent and membrane permeate (effluent). The air was fed into the reactor with a microbubble air diffuser, and airflow could be adjusted by an air flow meter. The retention times of wastewater in anaerobic and aerobic reactor were 3 h and 6 h respectively. In addition, ozonation of the excess sludge was carried out in a contact column with an inner diameter of 5 cm and a height of 1.2 m. Ozone was introduced into the column through a diffuser located at the bottom of the column, and the ozonated sludge was returned to the aerobic reactor through a pump. A schematic diagram of the process configuration is presented in Fig.1.

1.2 Experimental raw water and analytical methods

The test wastewater was collected from a septic tank in the second campus of Harbin Institute of Technology (HIT) and its characteristics are summarized in Table 1.

1.3 Analytical procedures

COD, BOD₅, NH₃-N, TN, TP, MLSS and MLVSS were determined using the standard methods issued by the Environmental Protection Agency (EPA) of China (1989). pH was measured using glass electrodes connected to a pH/ISE-3C pH meter. DO was detected with a YSI(MODEL 50B) dissolved oxygen meter.

2 Results and discussion

2.1 Biological treatment unit

The treatment system consists of biological unit and ozonation unit, and the whole test was divided into two stages. The biological treatment unit was run for over 2 months under controlled conditions to reach steady state. Then every stage lasted 2 months. At the first stage, only the biological treatment unit was run, and the experimental result is summarized in Table 2.
Table 2 Performance of all phases at the first experimental stage

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Raw water</th>
<th>Anaerobic effluent</th>
<th>Aerobic filtrate</th>
<th>Membrane permeate</th>
<th>Removal, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD, mg/L</td>
<td>237.74</td>
<td>113.57</td>
<td>37.58</td>
<td>21.35</td>
<td>91.02</td>
</tr>
<tr>
<td>BOD₅, mg/L</td>
<td>108.37</td>
<td>58.24</td>
<td>12.37</td>
<td>9.37</td>
<td>91.35</td>
</tr>
<tr>
<td>BOD₅/COD</td>
<td>0.456</td>
<td>0.513</td>
<td>0.33</td>
<td>0.439</td>
<td></td>
</tr>
<tr>
<td>SS, mg/L</td>
<td>76.3</td>
<td>28.64</td>
<td>-</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>NH₄-N, mg/L</td>
<td>24.37</td>
<td>24.68</td>
<td>1.57</td>
<td>1.24</td>
<td>94.91</td>
</tr>
<tr>
<td>TN, mg/L</td>
<td>28.84</td>
<td>25.12</td>
<td>4.67</td>
<td>4.41</td>
<td>84.7</td>
</tr>
</tbody>
</table>

2.1.1 Anaerobic phase

It can be seen that anaerobic process has removals of 52.23% and 46.26% for COD and BOD₅ respectively, and BOD₅/COD ratio was improved from 0.456 to 0.513, which supplied a good condition for post-positioned aerobic process. In addition, SS up to 62.5% was also removed because of hydrolysis and acidification in anaerobic reactor. In raw water, the ratio of soluble COD(SCOD) to total COD was 0.544 whereas it increased to 0.952 in anaerobic effluent. It could be implied that suspended organic matters were hydrolyzed to soluble state by hydrolysis bacteria, and the large molecule organics were biodegraded to small molecule organics, so the biodegradation ability of the anaerobic effluent was improved, which was more adapted to the post-positioned aerobic process. As to nitrogen, the ammonification of organic nitrogen resulted in a slight rise in NH₄-N concentration, and a little amount of TN loss in anaerobic phase was observed, this might be attributed to the formation of new anaerobic bacteria. At anaerobic phase, due to the existence of bio-carrier, the majority of bacteria was living in an attached state, only a small portion was in suspension. For there was no sludge wasted during experimental period, it is assumed that the anaerobic biofilm phase had a nearly zero excess production.

2.1.2 Aerobic phase

At this test stage, DO in aerobic tank was kept at a low level of about 0.8 mg/L and MLSS was maintained in the range of 3000—3500 mg/L. It was found from Table 1 that the MBR removed COD effectively, and an average COD removal of 81.20% for anaerobic effluent was achieved. Sometimes, the filtrate COD of the reactor (the mixed liquid samples collected were filtered by qualitative filter papers) had a big fluctuation, whereas COD of the membrane permeate was often kept at about 20 mg/L. In addition, due to the exclusion of the UF, MLSS and some large refractory organics was completely retained at aerobic reactor whereas some small molecule easy-biodegradable organics could penetrate UF and flowed out of membrane with permeate, so compared anaerobic filtrate with membrane permeate, it was noticed that the ratio of BOD₅/COD was increased from 0.33 to 0.439. It was also found that the system had a mean removal of 94.91% for NH₄-N, which indicated that a DO concentration around 0.8 mg/L was suitable to achieve a nitrification rate equal to the de-nitrification rate, and this result is in line with some previous findings (Münch, 1996). In aerobic MBR, due to the highly concentrated MLSS, there coexisted aerobic zone and anoxic zone, and simultaneous nitrification and de-nitrification could proceed in the presence of carbon source and low DO, and a total 84.7% of TN loss was achieved in the system. During this stage, the biological system had an average excess sludge yield of 0.213 (g MLSS/g COD removed).

2.2 Ozonation unit for excess sludge

At the second test stage, both sludge digestion and wastewater treatment were done simultaneously in the same treatment system. The schematic diagram is shown in Fig. 2.

The process consists of ozonation and biodegradation stages. The ozonation enhances the biological degradability of sludge to be treated. The ozonated sludge is then decomposed in the subsequent biological treatment. A part of the biomass is mineralized by biological treatment via ozonation. The degree of mineralization of the biomass became considerable with increasing biomass to be treated by the recirculation so that the mineralized biomass would be equivalent to that generated from organic substrate contained in the wastewater. It can be expected that the reactor produces only a minute quantity of excess sludge.

With an assumption that the mass mineralized at ozonation stage is negligible, then material balances
around biological treatment stage are formulated as follows:

\[
V \cdot \frac{dX_i}{dt} = YQ(\ell_i - L_n) - bVX_i - Q_nX_i - Q_nX_n + yQ_n(X_n + X_i), \tag{1}
\]

\[
V \cdot \frac{dX_i}{dt} = - Q_nX_i - Q_nX_i + b'Q_n(X_n + X_i). \tag{2}
\]

Under steady conditions, the left-hand sides of Eq. (1) and Eq. (2) are equal to zero, and these two equations would be reduced to the following form:

\[
Q_n / V = YN_i - b \cdot X_i / X - [1-(y+b')]Q_n / V. \tag{3}
\]

Where: \( V \) is the effective volume of biological reactor (L); \( X_n \) is the active biomass concentration in biological reactor (mg MLSS/L); \( X_i \) is the inactive biomass concentration in biological reactor (mg MLSS/L); \( Y \) is the sludge yield (g MLSS/g COD); \( y \) is the yield of ozonated sludge (g MLSS/g MLSS); \( b \) is the specific decay rate (d\(^{-1}\)); \( b' \) is the residual ratio of ozonated sludge to original excess sludge (g MLSS/g MLSS); \( Q \) is the influent flow rate (L/d); \( Q_n \) is the flow rate of the wasted excess sludge (L/d); \( Q_n \) is the flow rate of ozonated sludge (L/d); \( N_i \) is the organic loading rate (kg COD/(kg MLVSS·d)).

It can be seen from the Equation (3) that the sludge wasting rate is related to the parameters of \( Y, N_i, b, X_i/X, Q_n, \) and \( V \) and \([1-(y+b')]\). Once the biological treatment process and the quality of wastewater are determined, \( Y, N_i, V \) and \( b \) become constant, and the sludge wasting rate is dependent on \( X_i/X, Q_n \) and \([1-(y+b')]\). \( X_i/X \) represents the active component in sludge and \([1-(y+b')]\) denotes the biological mineralization of the ozonated sludge. It can be concluded that these two items are both influenced by the conditions of sludge ozonation. The higher the degree of sludge ozonation, the more the biological mineralization of ozonated sludge takes place, and \( X_i/X \) also can be maintained at a high level. Therefore, it is thought that the reduction of ozonated sludge is only influenced by the conditions of ozonation, i.e., ozone dosage and flow rate of ozonated sludge (\( Q_n \)).

### 2.2.1 Determination of ozone dosage

Batch studies were conducted to evaluate the effect of ozone dosage on the degree of cell lysis and hydrolysis. The following procedures were carried out in order to determine the ozone dosage for treating excess sludge: ozone gas was supplied at the bottom of the contact reactor, SCOD of the activated sludge were obtained at different reaction time. Then the plots of SCOD versus ozonation time were drawn. According to the ozonation, a suitable ozone dosage could reach the point at which cell could be liquefied and mineralized to a certain degree while the treatment cost for excess sludge should not be too expensive.

As can be seen from Fig. 3, SCOD increased as ozonation continued, reached a maximum around 2 h and stayed constant thereafter, whereas MLSS kept decreasing during 3 h of ozonation time. Therefore, it was concluded that ozonation results in cell lysis and the increase in SCOD was mainly due to cell lysis. After 2 h of ozonation, the amount of COD solubilized by ozone is equal to the amount of dissolved organics
oxidized by ozone, thereby resulting in no net change in the SCOD concentration. The data were obtained with an ozone gas concentration of 1.2 mg/L. The influence of ozone concentration on the mineralization and solubilization of sludge should be further investigated.

Based on the above experimental findings, 2 h was considered to be an appropriate ozonation time. Ozone concentration multiplies by ozonation time and air flow, and divides by MLSS of the sludge, then the ozone dosage of 0.1 kg O₃/kg MLSS was obtained.

### 2.2.2 The effect of ozonation on sludge yield

Combining biological treatment unit and ozonation unit together to reduce the excess sludge production. For there was no sludge withdrawal from the system, it was assumed that sludge yield was ratio of total generated sludge production to the organics removed.

\[ Y = \frac{V(MLSS_i - MLSS_f)}{QT(COD_{in} - COD_{eff})} \]  
(4)

Where \( MLSS \), is the sludge concentration of reactor at time \( t \) (mg/L); \( MLSS_i \), is the sludge concentration of reactor at time \( t \) (mg/L); \( COD_{in} \) is the COD of influent (mg/L); \( COD_{eff} \) is the COD of effluent (mg/L); \( T \) is the interval of sludge concentration measurement (d).

In anaerobic biofilm reactor, most of the microorganisms were fixed on support media with only a negligible amount of microorganisms in the bulk. In post-positioned aerobic MBR, sludge concentration was increasing with the prolongation of operation time. During the second experimental period of sludge ozonation, the effect of ozone dosage and flow rate of ozonated sludge on the sludge production was studied. It was found that the increasing rate of sludge production decreased with the increase of ozone dosage and flow rate of ozonated sludge. When the ozone dosage was 0.10 kgO₃/kgMLSS and flow rate of ozonated sludge was 0.02 Q, sludge concentration was kept at around 3200 mg/L and sludge yield was about zero. Increasing flow rate of ozonated sludge, a decrease of MLSS concentration in aerobic was observed.

### 2.2.3 The effluent quality of ozonation process

The operating parameters of biological unit at these two stages were almost identical. For ozonation unit, the two main parameters of ozone dosage and flow rate of ozonated sludge were controlled at 0.10 kgO₃/kgMLSS and 0.02 Q respectively, then the effect of sludge ozonation on effluent quality was also investigated, and the result is presented in Table 4.

### Table 3 The effect of ozone dosage and flow rate of ozonated sludge on sludge yield

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Flow rate of the ozonated sludge</th>
<th>Sludge yield, g MLSS/g COD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.01 Q</td>
<td>0.02 Q</td>
</tr>
<tr>
<td>0.07</td>
<td>0</td>
<td>-0.04</td>
</tr>
</tbody>
</table>

Note: ozone dosage was 0.10 kg O₃/kg MLSS

### Table 4 Performance of all phases at the second experimental stage

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Raw water</th>
<th>Anaerobic effluent</th>
<th>Aerobic filtrate</th>
<th>Membrane permeate</th>
<th>Removal, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD, mg/L</td>
<td>252.38</td>
<td>121.74</td>
<td>54.62</td>
<td>23.12</td>
<td>90.84</td>
</tr>
<tr>
<td>BOD, mg/L</td>
<td>119.34</td>
<td>68.41</td>
<td>12.74</td>
<td>10.23</td>
<td>91.43</td>
</tr>
<tr>
<td>BOD₅/COD</td>
<td>0.473</td>
<td>0.562</td>
<td>0.233</td>
<td>0.442</td>
<td>-</td>
</tr>
<tr>
<td>SS, mg/L</td>
<td>113.24</td>
<td>42.31</td>
<td>-</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>NH₃-N, mg/L</td>
<td>27.24</td>
<td>28.36</td>
<td>6.57</td>
<td>6.46</td>
<td>76.28</td>
</tr>
<tr>
<td>TN, mg/L</td>
<td>34.45</td>
<td>32.52</td>
<td>8.64</td>
<td>7.38</td>
<td>78.87</td>
</tr>
</tbody>
</table>

Compared with first experimental stage, it can be seen that COD of the aerobic filtrate had a considerable rise whereas UF could retain most large molecule organics in aerobic reactor and kept COD of UF permeate at a low value, it was also noticed that the inorganics had a certain accumulation in aeration tank, VSS/SS varied from 0.827 to 0.684 in two months. Due to sludge ozonation recirculation, some inert or refractory organics was returned to biological unit. Large molecular organics was retained in the
aerobic reactor by ultra-membrane whereas small molecular organics could penetrate through membrane and flowed out of reactor with effluent, so the removals for COD and BOD$_5$ are similar to that at first stage. But for the removal of nitrogen, there were some differences at these two experimental stages. At second stage, with the extension of sludge ozonation, more biomass than excess sludge was inactivated by ozone and was returned to biological tank in the form of organics, so sludge retention time (SRT) of the system was thus shortened. It was estimated that the SRTs of these two stages were 35 days and 12 days respectively, which had a little influence on organics removal, whereas nitrification was influenced significantly by ozonation, because the specific growth rate of *nitrobacteria* is lower than that of *heterobacteria*, so sludge ozonation resulted in a gradual reduction ratio of *nitrobacteria* in total biomass, and there was a rise of NH$_4$-$\text{N}$ and TN in UF permeate. Thus, in view of nitrification, the flow rate of ozonated sludge should be limited in a certain range. It was thought that applying aerobic biofilm process for activated sludge process is advantageous to ammonium removal, because most *nitrobacteria* is apt to growth on the support media, so ozonation of suspended biomass could not cause great loss to *nitrobacteria*.

### 3 Economic analyses

An electric fee of about 15 RMB Yuan is needed to produce 1 kg of ozone gas. Based on the experimental data, the operating cost of sludge ozonation could be calculated. A quantity of 0.02 $Q$ sludge with a 3200 mgMLSS/L of concentration was treated, and ozone dosage was 0.10 kg O$_3$/kg MLSS, so sludge ozonation operating cost is equal to (15 RMB Yuan/kg O$_3$ $\times 3.2$ kg MLSS/m$^3$ $\times 0.10$ kg O$_3$/kg MLSS) 4.8 RMB Yuan/m$^3$ sludge. For the flow rate of ozonated sludge was 0.02 $Q$, thus the additional ozonation cost for wastewater treatment is 4.8 RMB Yuan/m$^3$ ($\times (0.02/Q) = 0.096$ RMB Yuan/m$^3$ wastewater. Whereas, in a conventional wastewater treatment plant, the handling, treatment, and ultimate disposal of wasted biosolids accounts for from 50% to 60% of the operating costs of the plant(Brennan, 1999). Which means that sludge ozonation process could reduce the operating cost of wastewater and excess sludge treatment.

### 4 Conclusions

The whole test was divided into two stages, at first stage, combined anaerobic biofilm and MBR process was used to treat domestic wastewater, and average removals of 91.02%, 91.35%, 100%, 94.91% and 84.7% were achieved for COD, BOD$_5$, SS, NH$_4$-$\text{N}$ and TN respectively. In addition, a sludge yield of 0.213 (g MLSS/g COD removed) was obtained.

At second stage, ozonation was applied to get a minimization of sludge production, the ozone dosage and the flow rate of ozonated sludge were controlled at 0.10 kg O$_3$/kg MLSS and 0.02 $Q$ respectively. Under these conditions, the removal efficiency for organics was not affected but nitrification was influenced significantly. A zero sludge yield was achieved during the second experimental stage, and an additional ozonation operating cost for wastewater treatment is 0.096 RMB Yuan/m$^3$ wastewater.

### References:


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