Treatment and remediation of a wastewater lagoon using microelectrolysis and modified DAT/IAT methods

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Abstract

To examine treatment and remediation of a wastewater lagoon with poor biodegradability, a typical wastewater lagoon in Tianjin, China, was treated and remedied using microelectrolysis and modified demand aeration tank (DAT)/intermittent aeration tank (IAT) methods. After pretreatment by microelectrolysis, the removal efficiency of chemical oxygen demand (COD) was up to 64.6% and the ratio of BOC/COD in the effluent increased from 0.013 to 0.609. The removal rates of COD and NH4+-N were affected by sludge backflow rate, mixed liquor suspended solids (MLSS), and hydraulic retention time (HRT) in the modified DAT/IAT reactor. The highest removal rates of COD and NH4+-N were up to 78.9% and 62.6%, respectively, when the sludge backflow rate was 38.0 mL/min, the total HRT was 8.0 hr and MLSS was 4088.0 mg/L. In this case, some protozoa and metazoa were observed in activated sludge and biofilm carriers. Most of chrominance was removed by microelectrolysis treatment, while the modified DAT/IAT methods were more effective for COD and NH4+-N removal.

Key words: wastewater lagoon; biodegradability; microelectrolysis; DAT/IAT; ecological remediation

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Introduction

Global deterioration of water quality and water resources, along with people’s increasing requirements of high-quality water, has increased concern of and requirement for appropriate wastewater treatment technologies (Liu et al., 2006; Zhou et al., 2006; Kong et al., 2010). In recent years, the development of wastewater treatment technologies have greatly improved water quality and quantity and reduced the cost of treatment processes (Scholz and Fuchs, 2000; Zakour et al., 2001; Dhouib et al., 2003, 2006). However, treatment and remediation of some wastewater lagoons receiving highly toxic chemical industrial wastewater with poor biodegradability remains unsolved (Mariem et al., 2009). The ferrite process can be an effective technology in treating wastewater with poor degradability, but its expense has limited its remediation use (Montserratt et al., 2002; Lou et al., 2009). Additionally, although membrane bioreactors have been successfully applied to treat industrial wastewater over the past decade (Acharya et al., 2004; Kurian et al., 2006), the membranes can be defiled easily when used in mixed industrial wastewater treatment (Geng and Hall, 2006; Nghiem and Hawkes, 2007). Traditional activated sludge and its modified technology are inexpensive and widely used for wastewater treatment; however, many wastewater lagoons contain hazardous materials from industrial production that inhibit bioactivity of living organisms and reduce removal efficiency of toxic pollutants. In recent years, increasing attention has been paid to electrochemical methods for industrial wastewater treatment. Electrolysis, in particular, has been the most frequently used unit process because of its rapid reaction rate, high efficiency, and low sludge production. However, the high consumption of electricity limits its application and leads to the development of interior microelectrolysis in wastewater treatment (Jin et al., 2003).

Interior microelectrolysis (or iron chips filtration) has been used to pretreat industrial wastewater as it is highly adaptable to wide variations in wastewater composition (Yang et al., 2009). It has been studied for treatment of poorly biodegradable wastewater, including pesticide wastewater (Wu et al., 2003), pharmacy wastewater (Liu et al., 2005), and dye wastewater (Cheng et al., 2005). The iron and carbon materials cast into the container form numerous microscopic galvanic cells, while electrons...
are supplied from the galvanic corrosion of iron (anode) (Zhou and Fu, 2001; Cheng et al., 2007). Products released from the galvanic cell reaction include hydroxyl, atomic hydrogen and Fe(II), which have high activities to decompose contaminants (Li et al., 2004; Wang et al., 2008). In addition, some pollutants can also be removed by electrophoresis and adsorption (Han, 1991; Yang et al., 2009). Furthermore, interior microelectrolysis pretreatment is low-cost and effective because it does not require chemical coagulants and external power in the cases of coagulation and electrolysis (Xu et al., 2007).

The demand aeration tank (DAT) and intermittent aeration tank (IAT) are processes that possess the continuity of conventional activated sludge and the flexibility of the sequencing batch reactor (SBR) activated sludge process (Zhang and Wang, 1996; Zhang et al., 2004; Wang et al., 2007). Generally speaking, the influent and aeration of DAT is continuous. The effluent of DAT then flows into IAT through double guide walls and completes procedures such as aeration, sedimentation, decantation and discharge of excess activated sludge in the IAT process (Zhang et al., 2007).

According to the poor biodegradability and complexity of chemical industrial wastewater, a sequential system of microelectrolysis and modified DAT/IAT was proposed. Hazardous materials and pollutants with poor biodegradability can be eliminated by the preset microelectrolysis and the ratio of BOD/COD can be greatly improved so that microorganism activity cannot be restrained by hazardous materials in the wastewater. Other organic pollutants, ammonia nitrogen, and biodegradable pollutants can also be removed through DAT/IAT, and water quality can be greatly improved through the combined process.

As a cooperation project between the Chinese and Singapore Governments, the goal of “the Tianjin Eco-city” is to help mitigate global climate changes, strengthen environmental protection, save natural resources, reconcile energy conservation, and build a harmonious society. The Sino-Singapore Eco-city is located in Bin-Hai New District, a rapidly developing area in Tianjin, China, which has a 2.5–2.6 km$^2$ wastewater lagoon with an average depth of 2 m and a wastewater storage volume of about 5 million m$^3$. For over 30 years, the wastewater lagoon has accepted a great deal of household sewage and chemical industrial wastewater with poor biodegradability. Self-purification capacity has progressively deteriorated and the quality of water is worse than Class V. This lagoon is a threat to human health and hampers social and economical development of the Sino-Singapore Eco-city and it is important to develop and innovative economical and practical technologies to treat and remedy this wastewater lagoon.

1 Materials and methods

1.1 Raw wastewater

The raw wastewater samples used in this study were taken from a wastewater lagoon (Fig. 1) in the Sino-Singapore Eco-city, Tianjin, China. Analytical results indicated that COD, NH$^4_-$N, and other hazardous substances, including heavy metals and organic pollutants such as pesticides were the main contaminants of the lagoon. As the wastewater lagoon contains wastewater of varied composition and still accepts wastewater from surrounding industrial areas everyday, wastewater samples were used for the entire study. Water quality parameters are listed in Table 1.

1.2 Tested materials

Waste iron scraps were obtained from a mechanical processing plant in Tianjin, China. The iron scraps were immersed into a 10% NaOH solution for 10 min and soaked in 5% sulfuric acid for 20 min to remove surface oxidation and increase reaction activities. Finally, the scraps were cleaned two or three times with deionized water (Li et al., 2008). The granular activated carbon (GAC) with 0.65–1.04 mm particle size was also washed two or three times with water, and then soaked in water for 12 hr before use to eliminate GAC adsorption. The suspended biofilm carriers used in the modified DAT/IAT were bought from the Chunqiu Dingsheng Environmental Science and Technology Co., Ltd., Beijing, China.

1.3 Experimental equipments

The modified DAT/IAT experimental equipment without microelectrolysis pretreatment is shown in Fig. 2. The reactor was manufactured using 5 mm thick lucites. The volume of the hydrolytic acidification tank, DAT, and IAT with a protecting height of 80 mm was 100 mm × 200 mm.
x 300 mm, 280 mm × 200 mm × 300 mm, and 200 mm × 200 mm × 300 mm, respectively. The effective volume of DAT/IAT was 21.12 L. Because the wastewater samples contained some hazardous materials, several suspended biofilm carriers were put into the DAT reactor to promote their biodegradation.

Processed iron scraps and GAC at a certain proportion were put into a 1 L beaker. Then 800 mL of wastewater was poured into the beaker and pH was adjusted to a certain value by sulfuric acid. The beaker was stirred continuously with a mixer. Because of the high salinity (Table 1), the raw wastewater worked as an electrolyte and promoted microelectrolysis. After a certain time of mixing and reaction, the pH value was regulated to 8.5 with NaOH (Li et al., 2008). It was then aerated for 30 min. After sedimentation, the hydroxide precipitation was separated from the reaction system, and the supernatant flowed into the DAT process and then into the IAT process where biofilm carriers were placed in the modified DAT reactor to promote their biodegradation.

A proportion of sludge flowed back to the DAT process at a certain backflow rate, and the rest flowed out of the IAT process after sedimentation. The technological flow chart of the combined process is shown in Fig. 3 and the modified DAT/IAT experimental equipment with microelectrolysis pretreatment is shown in Fig. 4. The reactor of modified DAT/IAT experimental equipment with microelectrolysis pretreatment was exactly the same as that without microelectrolysis pretreatment, except the former had no hydrolytic acidification tank.

Fig. 2 Sketch of the modified DAT/IAT experimental equipment (without microelectrolysis pretreatment). (1) intake pump; (2) return-sludge pump; (3) aerator; (4) air blower; (5) air valve; (6) time controller; (7) electromagnetic valve; (8) mixer; (9) biofilm carrier.

Fig. 4 Modified DAT/IAT experimental equipment (with microelectrolysis pretreatment). (1) intake pump; (2) return-sludge pump; (3) aerator; (4) air blower; (5) air valve; (6) time controller; (7) electromagnetic valve; (8) biofilm carrier.

1.4 Experimental procedures

Activated sludge used in the reactor was collected from the TEDA Wastewater Treatment Plant in Tianjin, China. After 14-day culture and acclimation using wastewater samples from the wastewater lagoon, activated sludge and suspended biofilm carriers were placed in the modified DAT/IAT reactor without microelectrolysis. Water quality parameters including COD and NH$_4^+$-N in both influent and effluent were determined. In this stage, the operating parameters of DAT/IAT included sludge backflow rate at 38.8 mL/min, hydraulic retention time (HRT) at 12.0 hr, and mixed liquor suspended solids (MLSS) at 3383.0 mg/L.

The orthogonal method was used to optimize the experimental design of microelectrolysis, and influences on treatment efficiency of different parameters were investigated. The parameters chosen were pH in reaction beakers, volume ratio of Fe and reaction wastewater (V(Fe)/V), volume ratio of Fe and GAC (V(Fe)/V(C)), and mixing speed and HRT (Xu et al., 2003). Orthogonal experiment arrangement is listed in Table 2.

The modified DAT/IAT with microelectrolysis pretreatment experimental design and arrangement using L$_{16}$(4$^5$)

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<th>Factor c</th>
<th>Factor d</th>
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* L$_{16}$(4$^5$) means 5 factors with 4 levels. HRT: hydraulic retention time.

Fig. 3 Technological flow chart of the microelectrolysis and modified DAT/IAT joint process.
ment had two important stages. The first stage was start-up. After a 30-day culture and acclimation with effluent from microelectrolysis, wastewater samples were turned into clean water. In the meantime, the average removal efficiency of COD increased from less than 15% of the initial concentration to about 40% of the initial concentration and the effluent looked clear. The second stage was the operating stage, with operating parameters of DAT/IAT involved in 3 L of the influent per period pumped into the DAT reactor within 30 min, 3000–5500 mg/L of MLSS, and 5.0 mg/L of the average dissolved oxygen (DO).

1.5 Analytical methods

Water quality was analyzed according to the standard methods for Examination of Water and Wastewater (CEPA, 2002). Chroma, salinity, and COD were determined by the potassium dichromate method, the visual colorimetry, and a hand-held refractometer manufactured by the Xingchengguang Optical Instruments Factory (China), respectively. The parameters including DO, pH, biofacies, NH$_4^+$-N, and BOD were analyzed using a HQ-10 DO detecting instrument (HACH Company, USA) the PB-10 pH instrument (Sartorius Company), Leica DMIL fluorescence microscope (Leica Company), the nessler’s reagents spectrophotometer using a UV-Vis spectrophotometer (WD-9403D, Beijing, China), and a LY-05 rapid detection equipment (Lv Yu Environmental Protection Co., Ltd., China), respectively. The concentration of Pb was determined by the flame atomic absorption using a flame atomic absorption spectrophotometer (AA240FS, Varian, USA).

2 Results and discussion

2.1 Treatment efficiency of the modified DAT/IAT

After the activated sludge was placed into the modified DAT/IAT reactor, the operation of the reactor commenced. At the initial stage, it took some time for degrading microorganisms to adapt to the operating system. Therefore, removal efficiencies of COD and NH$_4^+$-N at the initial stage were low. As shown in Fig. 5, the removal efficiencies of both COD and NH$_4^+$-N by the modified DAT/IAT increased with time for the first 4 days during the adaptive period. When growth of degrading microorganisms stabilized, removal efficiencies of both COD and NH$_4^+$-N were subjected to the operating parameters and water quality. Conversely, COD and NH$_4^+$-N removal efficiencies decreased continuously after day 4. After that, the effluent turned a little yellow and foamy, accompanied with smaller sludge particles, sludge disintegration, and floats. From the microscopic examination of sludge, almost no protozoa or metazoan was found, except several rotifer bodies. There were many hazardous materials in the wastewater samples from the lagoon, including heavy metal ions and poorly biodegradable pollutants. With an increase in treatment time, heavy metals and other hazardous materials accumulated in the microbial bodies can also affect biological activity, biodegradation and longevity of microorganisms.

![Removal efficiency of COD and NH$_4^+$-N by the modified DAT/IAT.](image)

As a result, effluent water quality from the modified DAT/IAT worsened.

2.2 Optimum conditions of microelectrolysis

In principle, the operation of interior microelectrolysis is similar to electrochemical methods. Iron and carbon are applied as the anode and cathode, and electron flows are created by the galvanic cell instead of the external power supply (Yang, 2009). The half-cell reactions are represented as follows (Fan et al., 2009):

Anode (oxidation):

\[
2Fe - 4e^- \rightarrow 2Fe^{2+} \quad E^0(Fe^{2+}/Fe) = 0.44 \text{ V}
\]

Cathode (reduction):

\[
4H^+ + 4e^- \rightarrow 2H_2 \uparrow \quad \text{(Acidic)}
\]

\[E^0(H^+/H_2) = 0.00 \text{ V}\]

\[
O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad E^0(O_2) = 1.23 \text{ V}
\]

\[
O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \quad \text{(Neutral to alkaline)}
\]

\[E^0(O_2/OH^-) = 0.40 \text{ V}\]

In Table 3, $K_{ij}$ was the sum of removal efficiency of COD in the $i$ level ($i = 1, 2, 3, 4$) of $j$ factor ($j = a, b, c, d, e$) and $k_{ij}$ was the average value of $K_{ij}$. $R_j = max(k_{1j}, k_{2j}, k_{3j}, k_{4j}) - min(k_{1j}, k_{2j}, k_{3j}, k_{4j})$. The larger the $R_j$ value was, the larger the effect of various factors on the index, and the largest $R_j$ value should be the major influencing factor. From the orthogonal experiment, the $R_j$ value of 5 different factors on the removal efficiency of COD was $R_a (35.96) > R_d (13.08) > R_c (10.47) > R_b (9.32) > R_e (3.78)$. Thus, it can be inferred that the important sequence of these factors was $P_H > mixing speed > V(Fe)/V(C) > V(Fe)/V > HRT$.

The relationship between COD removal efficiency and orthogonal experimental conditions is depicted in Fig. 6. According to the preliminary analysis of the orthogonal experimental results, the optimum program was $a_1 b_4 c_4 d_2$ and $c_3$, and the corresponding conditions included: $pH = 2.0, V(Fe)/V = 0.03750$ (the volume of the influent was 800 mL), $V(Fe)/V(C) = 2.0$, mixing speed = 200 r/min, and HRT = 1.5 hr. Further verification showed that COD in effluent under optimum conditions was 121.6 mg/L and
### Table 3 Calculated results of orthogonal experiments using L₁₆ (4⁵)

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\[ \text{Max} (k) = 47.12 \]

Optimum condition a₁ b₄ c₄ d₂ e₃

* COD removal efficiency of the 4th orthogonal experiment (black font) was the best of the 16 orthogonal experiments.

* “∑” and “Average” mean total value and average value of COD removal efficiency, respectively.

### Fig. 6 Relationship between COD removal efficiency and orthogonal experimental conditions.

The mixing speed was also an important parameter in microelectrolysis. As mixing speed increased, pollutants reacted more often with reaction materials and degradation was accelerated. When the mixing speed was more than 200 r/min, flocculation of pollutants was scattered, and the removal efficiency of COD decreased.

As the value of V(Fe)/V(C) increased, the removal efficiency of COD improved because pollutants did not degrade adequately when the Fe concentration in the reactor was insufficient for forming enough primary cells. Superfluous Fe in the reactor may also generate a large amount of Fe²⁺ and Fe³⁺, however, and exist as Fe(OH)₂ and Fe(OH)₃ in basic medium. They can cover the surface of iron scraps and GAC, and inhibit the process of microelectrolysis. The same thing also occurs when HRT is too long.

### 2.3 Improvement of wastewater lagoon biodegradability by microelectrolysis

Changes in COD, BOD, NH₄⁺-N, and Pb in influent and effluent after microelectrolysis under optimum conditions are summarized in Table 4. Noticeably, removal efficiency of Pb was up to 31.6% after microelectrolysis. This led to lower effluent toxicity. Generally speaking, microelectrolysis aims to remove COD and NH₄⁺-N, with limited attention paid to removal of Pb. In this work, the higher removal efficiency of Pb was attained through special regulation of various parameters (Lucio et al., 2008; Wang et al., 2009). Conversely, it can be seen from Table 4 that NH₄⁺-N can be partly removed by microelectrolysis. The iron ions (Fe³⁺) generated in the electrode reaction enhanced activity of degrading microorganisms in the treatment unit of the follow-up bioprocess and changed...
the structure of sludge particles, which was good for the simultaneous action of nitrification and denitrification (Liu et al., 2008).

The BOD/COD ratio of wastewater was greatly improved, because Cl⁻ ions in the water were turned into Cl₂, Cl₂, and ClO⁻ in the electric field, resulting in indirect degradation of organic pollutants (Lichoung et al., 1995; Ribirdy, 1997). In addition, poorly biodegradable pollutants became easily biodegradable and toxic substances became less toxic through redox reactions, which were also probably responsible for the higher BOD/COD ratio in the effluent and provided a foundation for the follow-up bioprocess.

Some protozoa and metazoan such as vorticellas, ciliates, rotifers, and nematodes were observed in sewage sludge and suspended biofilm carriers of DAT/IAT through a microscope during the later stage of the combined process. Some microorganisms were immobilized on the carriers by the effects of covalent bonds and ionic bonds between the chemical groups on the carrier surface and the functional groups on the biological membranes. In practice, the carriers protected the immobilized microorganisms from shock at any sudden change in sludge backflow rate or HRT. The poison function also decreased by protection of the carriers, and microbial activities therefore improved. The experiment showed that the average sludge volume index (SVI) of DAT and IAT was 62.4 and the average SVI₉₀ was 27%, which indicated favorable biodegradation in the combined process.

### 2.4 Influences of sludge backflow rate from IAT to DAT

According to the above experimental method, the effect of sludge backflow rate from IAT to DAT on removal efficiencies of COD and NH₄⁺-N was investigated with an HRT of 12 hr (Fig. 7). When the backflow rate was 27 mL/min, the removal efficiencies of COD and NH₄⁺-N were both low. When it increased to 38 mL/min, the removal efficiencies reached the highest value during the experiments.

In the modified DAT/IAT, full degradation of toxic pollutants was gradually attained when sludge flowed back from IAT to DAT. Wastewater and sludge in the two reactors would not fully mix and the degradation of toxic pollutants and the nitration process was not adequate when the backflow rate was low. When the IAT reactor was in the sedimentation condition and a deep layer of sludge was in the anaerobic state, some macromolecules degraded into small molecules. In this sense, a proper sludge backflow rate from IAT to DAT can improve the exchange of pollutants between the anaerobic state and the aerobic state, and enhance the removal of pollutants. When IAT was in the sedimentary stage and wastewater flowed back to DAT, sludge accumulated alongside the sludge discharge pipe and the return-sludge pump brought back only wastewater. Wastewater from the DAT process to the IAT process contained a significant amount of sludge. The faster the backflow rate, the less MLSS in the DAT process; therefore, the removal efficiencies of COD and NH₄⁺-N both reduced when the backflow rate was faster than 38 mL/min. Additionally, when the sludge backflow rate was too fast, the retention time of wastewater in the DAT reactor was too short, and nitrification was also restricted.

#### 2.5 Influences of HRT and MLSS in the modified DAT/IAT

At the optimal sludge backflow rate (38 mL/min), the effect of HRT (HRT in the DAT reactor) on the removal efficiencies of COD and NH₄⁺-N were also investigated. In this investigation, HRT in the DAT reactor equaled that in the IAT reactor, and half the HRT in the IAT reactor was aeration and half sedimentation. Effluent flowed out of the IAT reactor through an electromagnetic valve for the last 30 min. The results (Fig. 8a) indicated that the removal efficiency of COD was up to 77.0% when HRT was 8 hr. When HRT either increased or decreased, the removal efficiency of COD decreased. Organic pollutants cannot be adequately degraded at a short HRT, but too long HRT caused self-degradation of microorganisms, thus COD in the effluent increases. It is clear that COD is the major removal indicator in this experiment, and HRT at 8 hr is recommended for the modified DAT/IAT reactor. Similarly, the removal efficiency of NH₄⁺-N reached 66.2%, the highest value when HRT was 6 hr. Generally speaking, HRT for nitrification is often 6 hr at least. Thus, the high removal efficiency of NH₄⁺-N can be attained with HRT.
longer than 6 hr.

Biological treatment of wastewater basically depends on the activity of degraded microorganisms in the sludge. In other words, MLSS can directly affect removal of organic pollutants. The effect of MLSS on the removal efficiencies of COD and NH$_4^+$-N by the modified DAT/IAT was also investigated according to the above experimental methods. The results indicated that highest COD and NH$_4^+$-N removal efficiencies were 78.9% and 62.6%, respectively, when MLSS was 4088 mg/L. As shown in Fig. 8b, too high or too low MLSS limited COD and NH$_4^+$-N removal. When MLSS was too low pollutants were not fully removed, when MLSS was too high degraded microorganisms received insufficient nutrition. Under such conditions, the self-degradation of microorganisms resulted in lower removal efficiencies of COD and NH$_4^+$-N.

Water quality was greatly improved after the combined process of microelectrolysis and modified DAT/IAT, with more major pollutants removed compared with only the DAT/IAT process. Thus, we concluded that the combined process of microelectrolysis and modified DAT/IAT was an effective method to improve water quality of lagoon wastewater containing mixed chemical industrial pollution.

### 3 Conclusions

In the modified DAT/IAT reactor without microelectrolysis pretreatment, microbial activities were seriously affected by heavy metals and other hazardous materials in the wastewater samples from the lagoon, which resulted in the continuous decrease in COD and NH$_4^+$-N removal efficiencies.

Microelectrolysis is a feasible method for the treatment and remediation of wastewater with low COD and high toxicity. After pretreatment, the removal efficiency of COD reached 64.6%, other hazardous materials were partly removed, and the BOD/COD ratio in the effluent increased from 0.013 to 0.609, which provided a good foundation for the follow-up bioprocess.

Optimal parameters for the modified DAT/IAT were a sludge backflow rate at 38 mL/min, HRT of DAT/IAT at 8 hr, and MLSS in the reactor at 4088 mg/L. Under the optimal conditions, microorganisms grew well in the reactor, the removal efficiencies of COD and NH$_4^+$-N reached 78.9% and 62.6%, respectively, and effluent water quality reached the state standard for water and wastewater analysis in China.

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