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Toxicity Reduction of Municipal Wastewater by Anaerobic-anoxic-oxic Process[1]

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Objective This study was conducted to optimize the operational parameters of anaerobic-anoxic-oxic (A^2/O) processes to reduce the toxicity of municipal wastewater and evaluate its ability to reduce toxicity. **Methods** A luminescent bacterium toxicity bioassay was employed to assess the toxicity of influent and effluent of each reactor in the A^2/O system. **Results** The optimum operational parameters for toxicity reduction were as follows: anaerobic hydraulic retention time (HRT) = 2.8 h, anoxic HRT = 2.8 h, aerobic HRT = 6.9 h, sludge retention time (SRT) = 15 days and internal recycle ratio (IRR) = 100%. An important toxicity reduction (%) was observed in the optimized A^2/O process, even when the toluene concentration of the influent was 120.7 mg L^{-1}. **Conclusions** The toxicity of municipal wastewater was reduced significantly during the A^2/O process. A^2/O process can be used for toxicity reduction of municipal wastewater under toxic-shock loading.

Key words: A^2/O process; Municipal wastewater; Toxicity reduction; Process optimization; Toxic-shock loading

INTRODUCTION

Assessment and optimization of wastewater treatment processes are often based on the pollutant removal efficiency. Wastewater treatment was originally developed with the primary goal of reducing odor in receiving waters; therefore, BOD_5 and SS were the primary evaluation indices. Later, in the 1970s, nitrogen and phosphorus removal were required to protect the receiving waters from eutrophication. As a result, many existing wastewater treatment facilities were upgraded to accomplish this goal. Among the available advanced biological wastewater treatment processes, A^2/O was often used for simultaneous nitrogen and phosphorus removal. Indeed, this method has been applied widely during the last three decades in municipal wastewater treatment plants (MWWTPs). With the development of reactor kinetics and a better knowledge of microbiology ecosystems, many advances in the removal of conventional items by A^2/O have been achieved[1-6]. However, few studies have focused on the ability of the A^2/O process to reduce the toxicity of municipal wastewater.

It is well known that municipal wastewater contains many types of toxicants with the potential to harm humans[7-11]. However, analyses for conventional indices, such as COD and single toxicant analysis, cannot enable a complete response to the potential adverse effects of wastewater on wildlife and humans due to the different interactions and biological activities found in municipal wastewater[12]. With the development of wastewater reuse, there has been increased community concern over the ecotoxicicology and environmental safety of wastewater. Thus, there has been great demand for evaluation of the ability of currently available wastewater treatment processes to reduce toxicity.

Accordingly, the present study was conducted to optimize the operational parameters of the A^2/O process and evaluate its toxicity reduction ability. The A^2/O process was conducted at the laboratory-scale

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to investigate the toxicity reduction potential in this system. The effects of the hydraulic retention time (HRT), sludge retention time (SRT) and internal recycle ratio (IRR) on both toxicity and nutrient removal were investigated. In addition, toluene was added to assess the toxic-shock resistance of the biological treatment process.

MATERIALS AND METHODS

Luminescence Bacterium Toxicity Bioassay

A 15-min standard luminescence bacterium toxicity bioassay was conducted according to the standard methods of national environmental protection agency (NEPA)[13]. The freeze-dried marine bacterium (*Photobacterium phosphoreum)* and the testing instrument (toxicity analyzer model DXY-2) were purchased from the Institute of Soil Science, Academic Sciences, Nanjing PRC. Toxicity was measured using the DXY-2 instrument by quantifying the decrease in light emission from the bacteria that occurred in response to exposure of the wastewater to 3% NaCl solution for 15 min. The luminescent bacterium inhibition rate (LBIR) has been shown to be an effective indicator of toxicity for all environmental samples[14-16]; therefore, it was used to express the toxicity of the wastewater in this study.

Chemical Analysis of Water Quality

Water quality items including water temperature, pH, DO, BOD_5, COD, SS, NH_4^+-N, TP, and TOC were measured according to the standard methods of the NEPA[13]. TOC was measured by a total carbon analyzer (TOC-Vcpn, Shimadzu Company). Gas chromatography (GC) was used for quantitative analysis of toluene. The GC detector used in the test was an Agilent 6 890N (30.0 m×0.32 mm×0.25 μm, HP-5 column, with N_2 as the carrier gas). The GC temperature program was 100 ℃ for 1 min, after which it was linearly ramped to 150 ℃ at 2 ℃/min, where it was held for 2 min.

Operational Conditions of the System

The system used in this study consisted of three plexiglass complete-mixed reactors in series. Specifically, the reactors consisted of an anaerobic reactor, anoxic reactor, and aerobic reactor with effective volumes of 7 L, 7 L, and 21 L, respectively.

The influents of the lab-scale A^2/O process were obtained from a local MWWTP in Shanghai, China. The seed sludge in the system was obtained from the returned sludge of an existing pilot-scale A^2/O process treating municipal wastewater. The entire system was operated in a room with a constant temperature of 25 ℃.

Calculations

The pollutant concentrations in anaerobic, anoxic, and aerobic reactors were calculated according to Eq. (1)-(3):

$$C_{il} = \frac{C_i + R \times C_{e1}}{1+R} \tag{1}$$

$$C_{i2} = \frac{(1+R)\times C_1 + r \times C_3}{1+R+r} \tag{2}$$

$$C_{i3} = C_2 \tag{3}$$

The overall pollutant removal efficiency of the A^2/O system was calculated according to Eq. (4).

Overall pollutant removal efficiency

$$(\%) = \frac{C_i \times Q - (Q-Q_w)\times C_e - Q_w \times C_3}{C_i \times Q} \times 100\% \tag{4}$$

The total percentage of pollutant removed from the influent was obtained by taking the sum of the percentage of the pollutants removed by the anaerobic, anoxic, aerobic and settling reactors. These percentages were calculated using Eq. (5)-(8), respectively:

%Anaerobic removal

$$= \frac{(C_{i1} - C_1)\times Q \times (1+R)}{(C_i - C_e)\times Q + (C_e - C_3)\times Q_w} \times 100\% \tag{5}$$

%Anoxic removal

$$= \frac{(C_{i1} - C_2)\times Q \times (1+R+r)}{(C_i - C_e)\times Q + (C_e - C_3)\times Q_w} \times 100\% \tag{6}$$

%Aerobic removal

$$= \frac{(C_{i3} - C_3)\times Q \times (1+R+r)}{(C_i - C_e)\times Q + (C_e - C_3)\times Q_w} \times 100\% \tag{7}$$

%Settling removal

$$= \frac{C_3 \times (Q + RQ - Q_w) - C_e \times (Q - Q_w) - RQC_{e1}}{(C_i - C_e)\times Q + (C_e - C_3)\times Q_w} \tag{8}$$

In the above equations, C_i is the pollutant (COD, TP, TN, TOC) concentration in the influent; C_1, C_2, C_3, Ce are the pollutant (COD, TP, TN, TOC) concentrations in the effluent from the anaerobic, anoxic, aerobic, and settling reactors, respectively; C_{e1} is the pollutant (COD, TP, TN, TOC) concentration in the return sludge; C_{i1}, C_{i2}, and C_{i3} are the pollutant (COD, TP, TN, TOC) concentrations in mixed liquors in the anaerobic, anoxic, and aerobic reactors, respectively. Q and Q_w are the influent flow rate and waste sludge flow rate, respectively. R and r are the recirculation ratios for activated sludge and mixed liquor, respectively.

RESULTS

When the system was operated steadily, the operational parameters were changed one by one in an attempt to determine the optimum operational conditions for reduction of the municipal wastewater toxicity. The returned sludge rate was maintained at 100% throughout the experiment.

The Effect of Hydraulic Retention Time (HRT) on Toxicity Reduction

Several studies have shown that HRT could have a remarkable effect on the adsorption, biotransformation and decomposition of pollutants in wastewater treatment plants[1, 17-18].

To investigate the effect of HRT on toxicity reduction, we kept the internal recycling ratio at 200% and increased the HRT by increasing the flow rate gradually. Six group experiments were conducted by increasing the HRT. The experimental results of the entire system are shown in Table 1.

TABLE 1

Treatment Performance of the A^2/O Process at Different HRT

n	Parameters	Performance	HRT of the Entire System, h					
			6.0	8.0	10.0	11.5	14.0	19.5
3	NH_4^+-N	Average Removal Efficiency, %	84.0	87.2	88.6	88.6	86.4	86.0
		Standard Deviation	3.4	2.6	4.1	0.6	3.0	1.0
3	COD	Average Removal Efficiency, %	91.0	90.0	90.4	90.6	92.0	92.7
		Standard Deviation	2.1	1.6	0.8	0.2	2.5	2.9
3	LBIR	Average Removal Efficiency, %	76.7	80.9	84.7	83.1	83.4	82.1
		Standard Deviation	4.9	1.6	0.3	0.9	1.9	1.6

TABLE 2

Effect of HRT on the Removal of COD and LBIR in the Anaerobic Reactor of the A^2/O System

n	Parameters	Performance	HRT of Anaerobic Reactor, h					
			1.2	1.6	2.0	2.3	2.8	3.9
3	COD	Average Removal Efficiency, %	53.9	53.2	60.5	62.6	62.6	59.9
		Standard Deviation	5.7	4.5	3.5	3.6	3.0	3.1
3	LBIR	Average Removal Efficiency, %	8.1	22.2	9.7	9.6	26.5	20.4
		Standard Deviation	0.5	2.0	1.4	1.3	2.5	3.5

TABLE 3

Effect of HRT on the Removal of COD and LBIR in the Anoxic Reactor of the A^2/O System

n	Parameters	Performance	HRT of Anoxic Reactor, h					
			1.2	1.6	2.0	2.3	2.8	3.9
3	COD	Average Removal Efficiency, %	2.8	9.2	8.4	10.7	6.9	5.3
		Standard Deviation	1.6	1.3	2.3	3.1	1.2	1.6
3	LBIR	Average Removal Efficiency, %	1.0	4.6	5.6	6.5	13.2	6.9
		Standard Deviation	0.7	2.3	1.3	3.6	2.8	2.2

TABLE 4

Effect of HRT on the Removal of COD and LBIR in the Aerobic Reactor of the A^2/O System

n	Parameters	Performance	HRT of Aerobic Reactor, h					
			3.6	4.8	6	6.9	8.4	11.7
3	COD	Average Removal Efficiency, %	24.9	22.2	28.7	27.8	28.9	28.1
		Standard Deviation	1.2	5.0	2.1	5.3	2.5	1.1
3	LBIR	Average Removal Efficiency, %	32.0	25.6	35.8	40.7	33.9	30.6
		Standard Deviation	0.6	2.8	4.4	0.4	2.1	2.8

As illustrated in Table 1, the change in HRT did not have a significant effect on the removal performance of the overall process. When the HRT of the entire system increased from 6h to 19.5 h, the average COD and NH_4^+-N removal efficiencies ranged from 89%-92% and 84%-88%, respectively. Additionally, the toxicity reduction efficiency reached a stable high level (above 80%) when the HRT of the entire system was greater than 8.0 h.

As shown in Table 2 - Table 4, the wastewater toxicity was primarily reduced in the aerobic reactor, where the LBIR reduction efficiency attained 40.7% at an aerobic HRT of 6.9 h. The highest LBIR reduction efficiency in the anoxic reactor was 13.2% at an anoxic HRT of 2.8 h, which was the lowest among the three reactors.

The Effect of Sludge Retention Time (SRT) on Toxicity Reduction

It was evident that a short SRT favored P-removal; however, a short SRT was not good for nitrogen removal because the generation time of nitrifying bacteria is long. Therefore, the SRT should be very carefully considered when designing a wastewater treatment system.

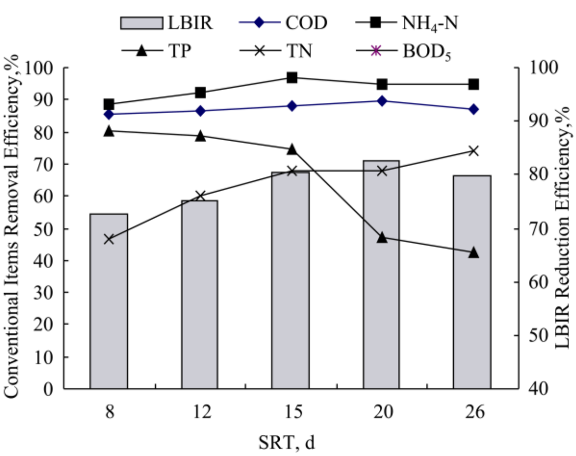

FIG. 1. Removal performance of the A^2/O process at different SRTs

During this stage, we maintained the internal recycling ratio at 200%, the anaerobic hydraulic retention time (HRT) at 2.8 h, the anoxic HRT at 2.8 h, the aerobic HRT at 6.9 h and gradually changed the SRT from 8 d to 26 d. As shown in Fig. 1, when the SRT increased from 8 d to 26 d, the COD and NH_4^+-N removal efficiencies were all greater than 85% and 82%, respectively. Additionally, the TN removal efficiencies increased from 46.9% to 74.2%, while the TP removal efficiencies dropped from 80.5% to 42.7%. It was evident that the removal efficiencies of TN and TP were severely influenced by the changes in SRT.

When the combined TN, TP and toxicity removal efficiencies were considered, an SRT for the system of 15 days was preferable. At this SRT, the COD, TN, TP and toxicity removal efficiencies were 88.2%, 68.0%, 74.6%, and 80.4%, respectively (Fig.1).

The Effect of Internal Recycle Ratio (IRR) on Toxicity Reduction

IRR values between 100% and 300% were recommended by Metcalf and Eddy for A^2/O processes treating municipal wastewater[19]. Four IRRs (0, 100%, 200%, 300%) were applied to A^2/O processes with an SRT of 15 days and a total HRT of 12.5 h. Mulkerrins found that the IRR increased from 100% to 300%, leading to improved NO_x-N removal in their system[20]. In agreement with their findings, a three-fold increase in IRR was found to lead to NO_x-N concentrations decreasing from 12.8 mg·L^{-1} to 9.4 mg·L^{-1} in the clarified effluents in the present study (Fig. 2).

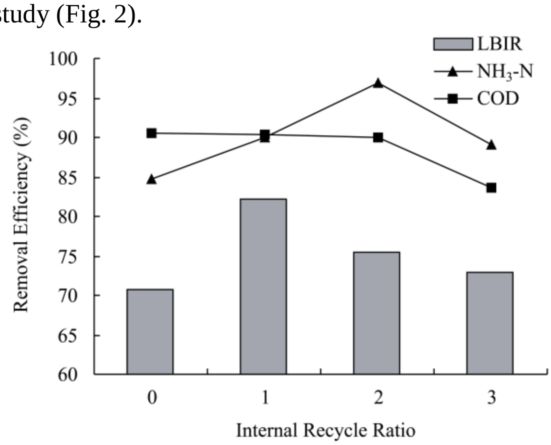

FIG. 2. Performance of A^2/O process at different IRRs

Performance of the A^2/O Process under Toluene Shock Loading

According to the results of our GC-MS analysis of the influents and effluents of several MWWTPs in Shanghai, China, toluene was widespread in the wastewater treated by these plants[21]. The toxic effects caused by the increase in the toluene concentration of the A^2/O system might lead to deterioration of sludge; therefore, toluene was selected as a typical toxicant in this study to analyze its impact on reduction of toxicity in the A^2/O process. The system was operated for two weeks for each selected toluene concentration. Fig. 3 shows the performance of the A^2/O process during the toxic shock loading experiment.

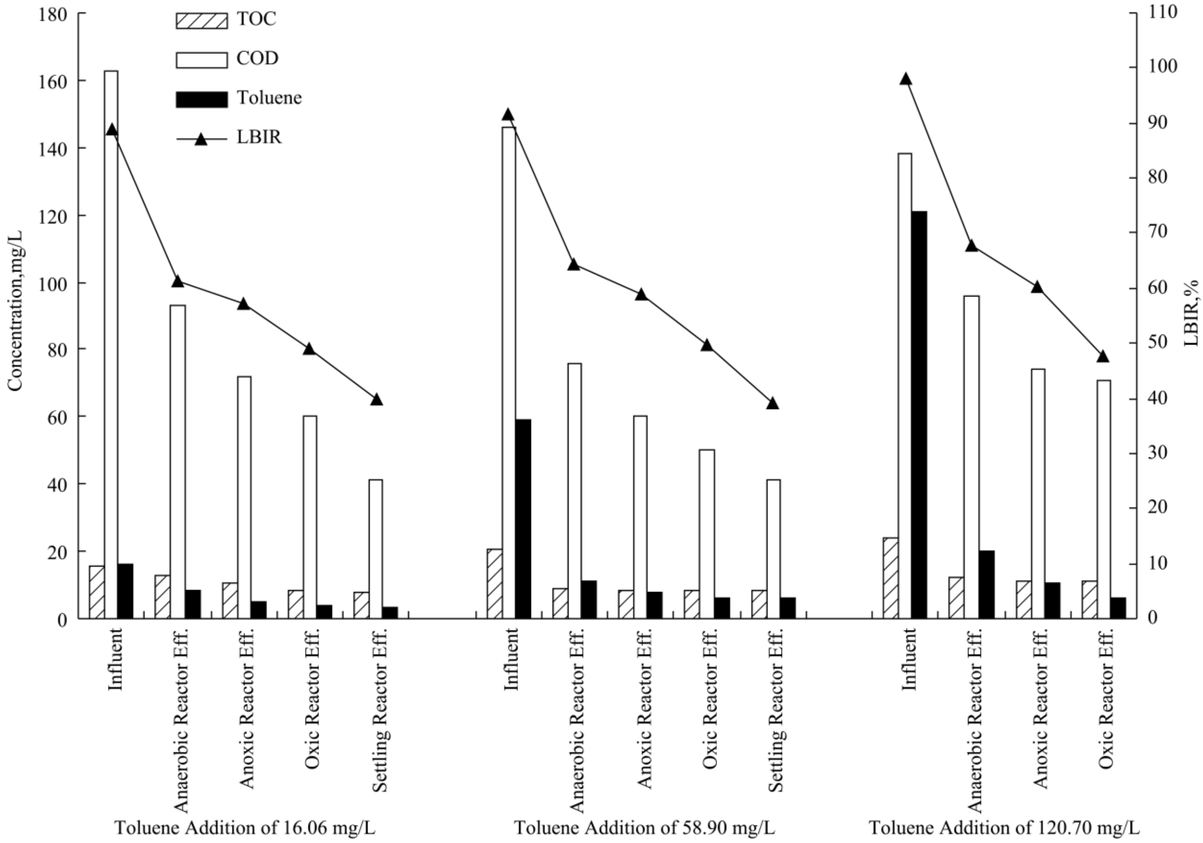

FIG. 3. Performance of A^2/O process after addition of toluene at different concentrations.

DISCUSSION

The Effect of Hydraulic Retention Time (HRT) on Toxicity Reduction

In the anaerobic reactor, more than 50% of the COD was removed, but the toxicity was reduced less in this reactor, with a maximum of 26.5% occurring at an anaerobic HRT of 2.8 h. This was likely because some of the refractory toxicants were decomposed and transformed into other types of toxicants in the anaerobic reactor. The highest LBIR reduction efficiency in the anoxic reactor was 13.2% at an anoxic HRT of 2.8 h, which was the lowest among the three reactors.

The Effect of Sludge Retention Time (SRT) on Toxicity Reduction

In the SRT range of 8-20 d, an increase in SRT was beneficial to toxicity reduction. Indeed, the toxicity reduction efficiency reached its maximum of 82.6% when the SRT was 20 d. However, when the SRT was longer than 20 d, the toxicity reduction efficiency decreased, which may have been the result of sludge decay and desorption of toxicants from the sludge.

The Effect of Internal Recycle Ratio (IRR) on Toxicity Reduction

Recycling of the mixed liquor from the aerobic reactor to the anoxic reactor was beneficial to the reduction of toxicity in wastewater. The toxicity reduction efficiency was highest at an IRR of 100%. A further increase of IRR may not help remove more toxicants. It was particularly evident that the toxicity reduction did not improve as the IRR was increased from 100% to 300%. In summary, the optimum operation conditions of the A^2/O process for toxicity reduction were as follows: HRT=12.5 h, SRT=15 d and IRR=100%. Under the above conditions, the removal ratios of NH_4^+-N, COD, BOD_5 and toxicity were 90.0%, 80.0%, 81.2%, and 82.2%, respectively.

Performance of the A^2/O Process under Toluene Shock Loading

As shown in Fig. 3, the A^2/O process could resist the toxic-shock loading caused by toluene.

Specifically, the addition of toluene at 16.06 mg/L, 58.90 mg/L, and 120.70 mg/L did not produce great disturbances in the system, and the toluene concentration of the effluent was very low when compared with that of the influents. Indeed, the system toxicity reduction efficiency was over 50%, even at a toluene influent concentration of 120.7 mg L^{-1}.

CONCLUSIONS

A laboratory scale experiment was conducted to evaluate the performance of the A^2/O process for reducing the toxicity of municipal wastewater. The results indicated that the A^2/O process could effectively reduce the toxicity of municipal wastewater in addition to COD and NH_4^+-N removal. The operational parameters of HRT, SRT and IRR had significant effects on the performance of the A^2/O process. Based on an experiment in which there was a constant returned sludge ratio of 100%, the optimum conditions of the A^2/O process for municipal wastewater toxicity reduction should be an SRT of 15 days, an anaerobic hydraulic retention time (HRT) of 2.8 h, anoxic HRT of 2.8 h, aerobic HRT of 6.9 h and IRR of 100%. Under the above conditions, the removal efficiencies of NH_4^+-N, COD and BOD_5 were 90.0%, 80.0%, and 81.2%, respectively. The *Luminescence bacterium* toxicity was reduced by 82.2%. The toxic-shock loading experiment revealed a significant decrease (%) in toxicity, even at a high inflow toluene concentration of 120.7 mg L^{-1}.

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