Original Research

Study on Anaerobic/Aerobic Membrane Bioreactor Treatment for Domestic Wastewater

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Abstract

A 69-day experimental study was conducted to investigate the treatment process of municipal wastewater using submerged membrane bioreactor technology. The results show that the average removal rate of COD and BOD₅ were over 90%. The removal rate of NH₃-N was 99% and the concentration of effluent NH₃-N was below 0.5mg/L; because of the high DO value in the denitration sect, the denitrobacteria was restrained and the effluent TN value was high, but the removal rate of TN can also reach 80%; the removal of TP was due to sample sludge and microorganism growth. The viscosity of sludge was not increasing with the sludge concentration, it was stabilized at 2.1 mPa·s. The sludge settling ratio (SV₃₀) and sludge volume index (SVI) were increased because soluble microbial products (SMP) and extracellular polymers (ECP) accumulated in the reactor during the course of the experiment, mixed liquor volatile suspended solids (MLVSS) and mixed liquor suspended solids (MLSS) fell slightly. The average diameter of sludge decreased in the course of the experiment, the diameter range was from 49.58 μ m to 24.96 μ m during the 69-day operation, and there was no trace of contamination membrane, illustrating that the PVDF flat sheet membrane manufactured by TORAY, Japan, had the ability to resist contamination.

Keywords: submerged membrane bioreactor, anaerobic/aerobic, sludge characteristics, domestic wastewater

Introduction

In view of the deteriorating condition of water pollution and water shortage in China, it is increasingly important to develop efficient technologies to treat contaminated wastewater. Some emerging treatment technologies, including membranous filtration, advanced oxidation processes, and the electrochemical method, hold great promises to provide alternatives for better protection of public health and environment.

In recent years, membrane bioreactor (MBR) technology has gained in popularity as a means of treating domestic wastewater, in part because of increasingly stringent dis-

Although MBR has many advantages, membrane fouling hinders its application tremendously. Membrane fouling not only results in reduced flux, which demands more frequent membrane cleaning, but also shortens the membrane life. Therefore, most research on MBR concentrates

charge requirements. Capable of producing high-quality effluent, the membrane units in these systems are almost always coupled aerobic reactors. Because nearly all bio-

mass can be retained within the reactor, MBR can maintain

a stable and high quality effluent. One more advantage of

MBR is that it can be operated with long solids retention

time (SRT), which is especially beneficial to slow-growing

microorganisms. As a result, decomposition of refractory

organic compounds can be achieved even under short

on controlling membrane fouling.

hydraulic retention time (HRT).

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The main advantage of using MBR technology over other conventional biological processes is to produce quality water from municipal wastewater for reuse, meeting the need for saving water, particularly in regions of water shortage. Other advantages include the need for less space, lower energy consumption, and the smaller excess of sludge to be handled. All shortcomings of membrane systems persist in MBR applications, such as high installation costs, low permeate flux, and the occurrence of membrane degrading and fouling. Despite performing excellently over years of full-scale operation, the interactions between microbes and the membrane in the MBR process, which determines its design and operational criteria, remain unclear. Just recently some mathematical modeling works were available for MBR applications [1-7].

The purpose of this paper is to explore and describe wastewater treatment using a membrane bioreactor system with microfiltration (UF), especially to investigate its applicability in the domestic wastewater treatment.

Materials and Methods

Reactor and Operation

The fully automatic submerged membrane bioreactor used in the experiment was designed and constructed jointly by Dalian Municipal Design and Research Institute of Environmental Science, Liaoning University of Petroleum and Chemical Technology and Tokyo University. The schematic diagram of the process flow and pilot plant setup was shown in Fig. 1. Influent (raw water) was transferred by a submersible pump to a feed influent tank through a strainer, which retained and prevented large suspended

solids from clogging the membrane. From the feed water tank, feed water was pumped to a slantwise sheet sedimentation tank and overflowed into a preposition denitrification anaerobic section (A), and then into a postposition aerobic section (O). In the final stage a membrane filtration pump (P3 as in Fig. 1) withdraws clear water through the submerged membranes in the aerobic tank at a flow rate of 42L/hr. The membrane filtration pump ran intermittently (13 min ON/2 min OFF) at operating pressure of 0-50kPa (zero means shutdown and 50kPa means the membrane runs under normal conditions). The aerobic tank was aerated continuously. NaClO was added by a pump (P4 as in Fig. 1). Capacities of all reactor tanks are indicated in Fig. 1. The membrane module used was Polyvinylidene Fluoride (PVDF) flat sheet membrane, a product from TORAY, Japan. The membrane had a pore size of 0.08 µm, a porosity of 70% and an effective filtration area of 0.45m². Fouling of the MBR membrane was prevented by providing intense aeration and intermittent effluent discharge (13 min ON/2 min OFF). A 4-section membrane module of high durability and large flow capacity was used in the experiment. Each section of the membrane module could be removed easily for maintenance and cleaning.

Wastewater Characteristics

Characteristics of wastewater are tabulated in Table 1.

Analytical Methods

To monitor the performance of reactors, samples of influent and effluent were analyzed for selected parameters using qualified instruments and procedures. pH value was

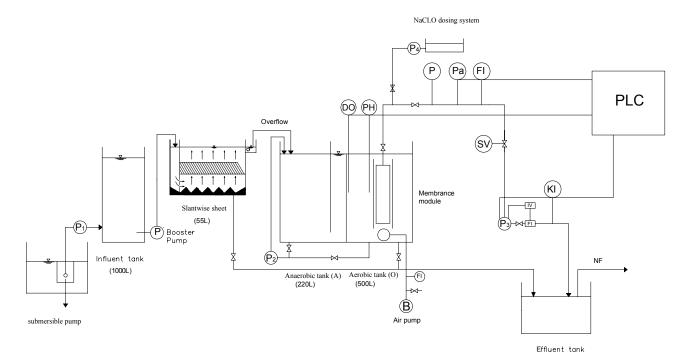


Fig. 1. Schematic diagram of A/O MBR for wastewater treatment.

Table 1. Characteristics of wastewater.

Temperature °C	Turbidity NTU	PH	COD mg·L ⁻¹	BOD ₅ mg·L ⁻¹	NH ₃ -N mg·L ⁻¹	TN mg·L ⁻¹	TP mg·L ⁻¹
13-17	150-196	7.0-8.1	230.85-630.17	119-367	29.84-75.1	40.67-88.97	3.03-6.49

Table 2. Analytical method of experimental wastewater quality.

Item	Method			
turbidity	Turbidity meter (Model 8391-37 Turbidity, USA)			
pН	pH meter (pHs-3C, China)			
COD	CIL-12 COD meter (Huatong Company, China)			
BOD ₅	BODTrakTM (Hach Company, USA)			
NH ₃ -N	Nesster's reagent colorimetry			
TN	Potassium persulfate dispels ultraviolet spectrophotometric method			
TP	Ammonium molybdate spectrophotometric method			
MLSS	weight measurement method			
MLVSS	weight measurement method			
viscosity	Viscosity meter (NDJ-79 viscosity meter, China)			

measured by a pH meter (pHs-3C, China). Chemical oxygen demand (COD) was measured by a CIL-12 COD meter (Huatong Company, China). Five-day biochemical oxygen demand (BOD₅) was determined by a BODTrakTM (Hach Company, USA). Turbidity was measured by a turbidity meter (Model 8391-37 Turbidity, USA). Dissolved oxygen (DO) and temperature were registered with a portable DO meter combined with a temperature probe (JBP-607 DO, China). Ammonia nitrogen (NH₃-N), color, suspended colids (SS), total suspended solids (TSS), oil, and total phosphor (TP) was conducted by the following methods. The analytical methods are presented in Table 2.

Results and Comments

COD and BOD, Removal

The inoculate sludge was taken from the Third Petroleum Factory of Fu Shun water treatment plant. After the acclimation of sludge, we put it into reactor and ran the system. During the experiment, influent COD stabilized between 230.85mg/L and 653.17 mg·L⁻¹. As shown in Fig. 2, the effluent COD was stable mostly between 10 mg·L⁻¹ and 20 mg·L⁻¹. This showed that the system could provide and maintain a steady effluent water quality whether the influent COD concentrates were high or low. This just shows that the system was capable of handling impact load condition. We also can see from Fig. 2 that active sludge

has the main function for COD removal, and membrane has the main function for maintaining stable system effluent.

Effluent BOD₅ was lower. As shown in Fig. 3, the average BOD₅ removal rate achieved 90.9%. That means membrane systems had a good result for BOD₅.

NH₃-N Removal

In order to study the elimination effect of NH_3 -N in the membrane bioreactor, the experimental operating conditions are: DO value control above 4.5 mg/L, influent pH value of $7.0\sim8.1$, reactor temperature of $13^{\circ}\text{C}\sim17^{\circ}\text{C}$, HRT at 17.14 h. The elimination effect of NH_3 -N in the membrane bioreactor was shown in Fig. 4. As shown in the chart,

- (1) in the first 15d, the NH₃-N elimination effect was quite bad; the elimination rate reaches 77.12% slowly. On the one hand, this was because the influent ammonia nitrogen load was big, but the nitrifying bacteria generation time was long, so the multiplication speed was slow. On the other hand, because the pH of the aerobic process was 4.65~5.8 and had not performed to control, low pH had an inhibitory action on ammonia nitrogen degeneration. In the latter 15d, the NH₃-N elimination rate rose above 99%, because sodium bicarbonate added to the aerobic process controlled the pH in the 6.5~7.0 range, enabling it to achieve suitable pH scope for nitrogen removal.
- (2) The value of NH₃-N removal by membrane was low, indicating that the elimination of NH₃-N depends on microorganisms in the membrane bioreactor; the reason for the membrane module's limited interception function was the small molecular weight of NH₃-N.

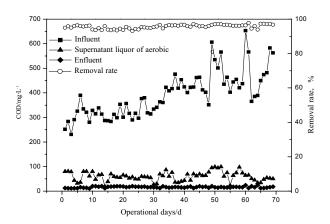


Fig. 2. Influent and effluent COD changed with time.

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(3) In the membrane bioreactor, the sludge residence time was long, made the generation time long, allowing nitrifying bacteria to accumulate gradually in the system; simultaneously, dissolved oxygen was high, enabling the full nitration of NH₃-N in the wastewater where the effluent stabilized below 0.5 mg/L.

TN Removal

As shown in Fig. 5, effluent TN value was high because in the reactor, in order to ensure nitrifying bacteria's nitration conditions as well as appropriate membrane surface washout intensity, the returned sludge leads the high DO value of the anerobic process, causing denitrifying bacteria suppression. In the membrane bioreactor, the average removal rate of TN can rise above 80%, because the membrane bioreactor apparatus does not carry a row of sludge beside the sample. The assimilation of sewage ammonia nitrogen and of organic nitrogen also enables TN to achieve certain levels of elimination.

TP Removal

The total phosphorus elimination effect in the membrane bioreactor was shown in Fig. 4. In this experiment, besides the sample, a row of sludge was not carried; theoretically speaking, the phosphorus elimination effect was not present. But in the experiment it was discovered that the phosphorus level in effluent was reduced. The reason mainly seems to be the partial phosporus as carried over along with the sample sludge. In conventional active sludge systems, phosphoric quantities of active sludge lie generally for dry weight between 1.5%~2.3% and with normal microorganism growth. In the membrane bioreactor, the nutrition from which the microorganism grows was more similar to that of conventional active sludge BOD₅:TN:TP=100:5:1. Therefore the microorganism may remove the partial nitrogen and the phosphorus during degeneration of the organic pollutant. In this experiment the phosphorus elimination was mainly caused by microorganism growth. As shown in Fig. 4, the membrane cut-off rate of TP may reach 19.43% because the membrane filtration function caused the phosphate, which was difficult to dissolve; the phosphorus and the calcium magnesium ion production settling was kept in the reactor, thus strengthening total phosphorus elimination.

Concentration and Viscosity of Active Sludge

In the beginning, the sludge concentration noticeably declined from an initial level of 6 g/L to 5 g/L. In accordance with Hrudey's [8] conclusions, nitration efficiency was not influenced by the decline of sludge concentration, which can be presumed that the decline of the sludge concentration was caused by heterotrophic bacteria, which did not survive in the membrane bio-reactor. The sludge concentration increased with the system running, and ultimately increased to 8.14 g/L.

Sludge viscosity had the same trend as the sludge concentration at startup, from 4 mPa/s down to 3 mPa/s. However, viscosity did not increase with an increase in sludge concentration. It basically stabilized at about 2.1mPa/s during the experimental period. This minute decreasing trend in viscosity demonstrated that the viscosity of sludge with more heterotrophic bacteria was higher than the viscosity of sludge with more nitrification bacteria.

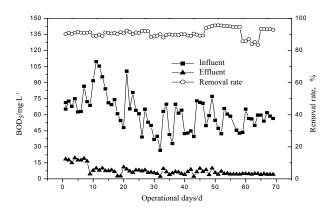


Fig. 3. Influent and Effluent BOD₅ Changes with Time.

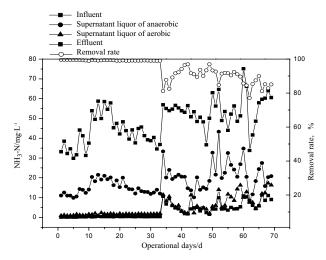


Fig. 4. Influent and effluent ammonium changed with time.

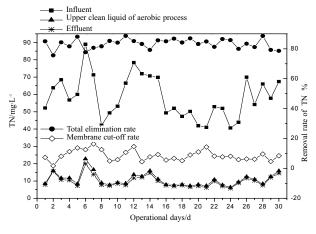


Fig. 5. Influent and effluent TN changed with time.

Moreover, viscosity slightly increased during the experimental period. The main reason could be related to the differences in extracellular polymer composition and concentration.

Sedimentation Capability of Active Sludge

As shown in Fig. 7, during the first 33 days of the experimental period, the sedimentation of sludge was good, with average SV_{30} and SVI at 35.35 percent and 60.72 ml/g, respectively. During days 34 through 44, due to impact loading of the influent, the SVI abnormally increased to 147.63 ml/g. However, SVI progressively declined with the stabilization of the system.

During the final 30 days of the experiment the increase in sludge load and concentration caused poor sedimentation and the average SV_{30} and SVI were 94.46 percent and 127.45 ml/g, respectively. After the experimental period, the bacteria's autolysis produced a great deal of soluble microbial products and extracellular polymers. The unique operational characteristic of the MBR caused soluble microbial products and extracellular polymers to accumulate in the reactors, resulting in poor sedimentation capacity and increased SV_{30} and SVI values.

In traditional active sludge technology, poor sedimentation capacity can result in poor effluent quality. In the experiment, because the MBR separated water from sediment, the system operation was not affected by poor sedimentation.

 SV_{30} and SVI were increased because the soluble microbial products (SMP) and extracellular polymers (ECP) accumulated in the reactor during the course of the experiment. We attributed the irregular accumulation of the biomass to variations in the biodegradability of SMP and ECP components [9].

Activity of Active Sludge

As MBR technology offers longer sludge retention time, higher sludge concentration and lower sludge load characteristics, sludge activity has become a focal point. There are many factors influencing sludge activity. Other than the basic process parameters, additional factors include membrane separation for inorganic matter, accumulation of refractory organic biomacromolecules, dissolved product of micro-organisms in the reactor, etc. The description of sludge activity was extensive as well. The simplest representation was to examine the ratio of volatile solids to total solids, i.e. MLVSS/MLSS. These ratios were used to express sludge activity in the test.

During the whole experimental process, the sludge activity, expressed in MLVSS/MLSS, had a declining trend that was particularly distinct in the end; the activity reflected the ratio of inorganic components in the sludge. Segregation of the influent's inorganic suspension by the reactor increased suspended solids and decreased the MLVSS/MLSS ratio. Judging from the overall operating

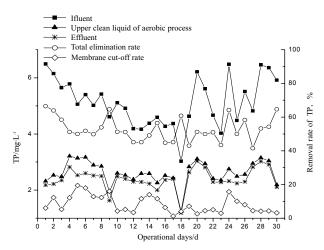


Fig. 6. Influent and effluent TP changed with time.

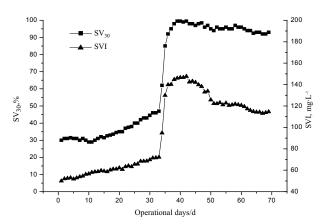


Fig. 7. SV₃₀ and SVI changed with time.

process, the ratio change was small, at an average of 0.69; the average MLVSS/MLSS ratio of inoculated sludge was 0.71.

Grain Diameter Distribution of Active Sludge

The experiment also analyzed the distribution of active sludge's grain diameters. Grain diameter distribution was tested using a laser instrument that utilized a light-scattering principle to collect and handle different ranges of grain diameters. The average grain diameter of inoculated sludge was found to be 49.58 μm . After system operation, the grain diameters decreased from 49.58 μm to 24.96 μm .

The reasons for decreasing grain diameters were a longer sludge retention time in the MBR, and the destruction of polymerization noted as a netlike sludge flocculation structure caused by the aeration system, which prevented membrane fouling and provided enough dissolved oxygen for microorganisms in the aerobic tank. The sludge flocculation from microorganisms, inorganic pellets, extracellular polymers and positive ions released from the extracellular polymers resulted in smaller sludge grain diameters.

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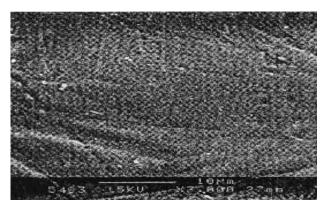


Fig. 8. Outer surface of new membrane.

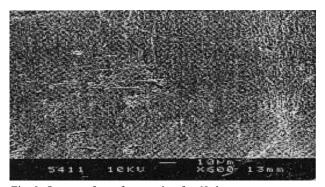


Fig. 9. Outer surface after running for 69 days.

Scan Electro Microscope (SEM) Pictures

Figs. 8 and 9 are SEM pictures for the membrane when it was new and after use in the pilot. From the pictures we can see no fouling was found in the membrane during the operational period of 69 days.

Conclusions

- 1 The average removal rates for COD and BOD₅ were found to be more than 90%. The removal was mainly done in the bioreactor area. The results showed that MBR was capable of handling impact load conditions for COD and BOD₅.
- 2 The NH₃-N elimination rate in A/O integration membrane bioreactor may reach above 99%, the effluent stabilizes basically below 0.5mg/L, but the nitrifying bacteria was influenced by remarkable pH; if you want to achieve the ideal elimination effect you must control the pH of the aerobic process. This experiment added sodium bicarbonate to control pH of the aerobic process.
- 3. Average removal rate of TN can rise above 80%, because the membrane bioreactor apparatus does not carry a row of sludge besides the sample. The assimilation of sewage ammonia nitrogen and of organic nitrogen also enables TN to achieve certain levels of elimination.

- 4. This A/O integration membrane bioreactor does not carry on a row of sludge, but also has the certain extent elimination of TP, this was because the sample sludge has carried over the partial phosphorus as well as satisfies microorganism growth to consume the partial phosphorus.
- 5. Under normal conditions the viscosity did not increase with the increase in sludge concentration, stabilizing at 2.1 mPa/s.
- 6. With the system running, bacteria autolysis produced a great deal of soluble microbial products and extracellular polymers. Moreover, the characteristics of the MBR made soluble microbial products (SMP) and extracellular polymers (ECP) accumulate in the reactor, which caused poor sedimentation capability and high SV₃₀ and SVI values. Average SV₃₀ and SVI increased from 35.35 percent and 60.72 mL/g, to 94.46 percent and 127.45 ml/g, respectively.
- 7. The activity expressed by MLVSS/MLSS reflected inorganic component in the sludge Increased membrane suspended solids resulted in a decline in these ratios from 0.71 to 0.69. Because of long sludge retention time and the aeration system in the aerobic tank, the grain diameter fell from 49.58 µm to 24.96 µm
- 8. No fouling was found in the membrane during the operational period of 69 days, this was clear in the SEM pictures taken for the tested membrane.

Acknowledgments

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