Wei KANG^{1*}, Xiyu CUI¹, Yanrui CUI¹, Linlin BAO¹ and Kaili MA¹

ASSESSMENT OF HIGH SALINITY WASTEWATER TREATMENT WITH DEWATERED ALUM SLUDGE-AEROBIC MEMBRANE REACTOR

Abstract: The discharge of wastewater containing both high salinity and high organic content without prior treatment is detrimental to aquatic life and water hygiene. In order to integrate the advantages of membrane treatment and biological treatment, and exert the phosphorus removal efficiency of dewatered alum sludge, in this study, an aerobic membrane reactor based on dehydrated alum sludge was used to treat mustard tuber wastewater with salinity of 6.8-7.3 % under the conditions of 30 °C, 20 kPa trans-membrane pressure (TMP) and chemical oxygen demand (COD) of 3300-3900 mg/L. Three replicate reactors were applied to assess the operational performance under different organic loading rate (OLR). The results showed that all reactors were effective in removing COD, ammonia nitrogen ($\text{NH}_4^+\text{-N}$) and soluble phosphate (SP) under the conditions of 30 °C and 20 kPa of TMP. Meanwhile, the effluent concentration of COD, $\text{NH}_4^+\text{-N}$ and SP all increased while OLR was changed from 1.0 to 3.0 kg COD/m³/day, and the effluent COD and $\text{NH}_4^+\text{-N}$ concentration except for SP could reach the B-level of Chinese “Wastewater quality standards for discharge to municipal sewers” when OLR was less than 3.0 kg COD/m³/day. This indicates that dewatered alum sludge-based aerobic membrane reactor is a promising bio-measure for treating high salinity wastewater.

Keywords: organic loading rate, biological treatment, membrane treatment, high salinity wastewater, trans-membrane pressure

Introduction

Fuling District, Chongqing is famous for its mustard tuber production market. During the manufacturing process, large amount of wastewater containing 0.3-20 g/L organic compounds and 0.5-7 g/L suspended solid are produced. Meanwhile, the wastewater is also detected with high salinity of 2-15 %, which will inevitably inhibit the biological treatment efficiency, considering the following aspects: (1) high salinity inhibits the activity of many enzymes in cell, and ultimately lead to plasmolysis [1, 2]; (2) high salt in wastewater affects anaerobic ammonium oxidation, nitrification and denitrification [3-5]; (3) high salinity reduces dissolved organic carbon (DOC), soluble phosphate and biochemical oxygen demandremoval rate [6-8].

In order to solve the above problems, a lot of researches on biological treatment technology have been carried out in recent years. Gaoet al. [9] adopted a system combining an anaerobic fluidised bed (AFB) with microbial fuel cell (MFC) to treat high salinity

¹ Key Laboratory of Yellow River and Huai River Water Environmental and Pollution Control, Ministry of Education, Henan Normal University, 453007, China, phone +86-18323216594, ORCID: WK 0000-0003-1750-6799, XC 0000-0001-9437-1360, YC 0000-0002-2331-8527, LB 0000-0002-5730-8909, KM 0000-0003-4858-6429

*Corresponding author: loulantingxue@126.com

wastewater, and the anode attained good removal efficiency of 98.6 % for chemical oxygen demand (COD) and 52.1 % for ammonia nitrogen ($\text{NH}_4^+\text{-N}$) when the Cl^- concentration was $< 6500 \text{ mg/L}$. A cyclic activated sludge system (a new technology of sequencing batch reactor (SBR) process evolution) was used to treat mixed mustard wastewater and urban sewage, and all indicators were stable to meet urban sewage treatment plant pollutant discharge standards when mixing ratio was less than $30 \pm 1 \%$ [10]. Considering the simple structure and low operation cost, SBR was used to treat high salinity wastewater by Tatoulis et al. [11] and Deorsola et al. [12], and removal rate of dissolved COD reached 96.5 % when initial COD concentration and operating mode of trickling filter were 7500 mg/L and SBR with recirculation, respectively. Considering upflow anaerobic sludge blanket (UASB) process has various advantages such as high efficiency and withstanding organic shock loading, a laboratory scale UASB reactor was adopted to treat synthetic wastewater at a hydraulic retention time (HRT) of 24 h, finally achieving a 88 % total organic carbon (TOC) removal efficiency under 2 % salinity and $1 \text{ g/L NH}_4^+\text{-N}$ [13, 14].

For better treating high salinity wastewater, membrane technology is also introduced and integrated into biological treatment. Lu et al. [15] adopted sequencing batch biofilm reactor to treat synthetic wastewater containing 14 g/L salinity, and the removal efficiency of COD reached 88.0 % at an OLR of $0.96 \text{ kg COD/m}^3\text{/day}$. As membrane bioreactor (MBR) has greater biomass retention time and less sludge production than conventional aerobic reactor, Yang et al. [16] and Dan et al. [17] used different MBRs to treat high salinity wastewater, and the maximum COD removal rate of yeast membrane bioreactor reached $0.93 \text{ g COD/g MLSS/day}$ at F/M of $1.5 \text{ g COD/g MLSS/day}$. Considering membrane fouling problems, many hybrid MBRs have been developed to eliminate this drawback [18]. A forward osmosis membrane bioreactor with thin film composite membrane and seeded with flocculated sludge or aerobic granules was used to treat synthetic wastewater with 1 M NaCl draw solution. The result showed the removal efficiencies of TOC, $\text{NH}_4^+\text{-N}$ and phosphate were 100, 43.0 and 96.0 %, respectively [19]. Patsios et al. [20] used a moving bed biofilm reactor-membrane bioreactor (MBBR-MBR) to treat real high-strength, saline wastewater from a table olive processing plant under three different HRTs of 88, 96 and 104 h, and the removal efficiencies of TOC and total polyphenols (TPH) were 85.6 and 89.1 %, respectively.

For a long time, the exploration of low-cost adsorbent and its application in wastewater treatment have been the focus of attention among by-products or wastes in various industries. Waterworks sludge is a by-product of coagulation, flocculation and other water treatment processes in drinking water treatment plants, which composition is complex, and the waterworks sludge is rich in aluminium, iron, calcium and other elements. To a certain extent, it is a waste of resources to bury it or throw it into the sea. Alum sludge has strong predictive adsorption capacity for phosphorus and is higher than most other typical low-cost adsorbents [21, 22]. Zhao et al. [23] and Yang et al. [24] integrated the alum sludge into a constructed wetland system for P-rich wastewater treatment. The results demonstrated that alum sludge can increase the phosphorus removal efficiency, and active bacteria attached growth on the alum sludge can effectively remove organic matter and $\text{NH}_4^+\text{-N}$. In addition, there are studies to indicate that P-adsorption capacity would be reduced with the increase of alum sludge particle sizes, and P recovery after the alum sludge reuse is feasible from a technical point of view [25].

Many studies have shown that the membrane fouling in anaerobic membrane bioreactor was more intense than aerobic membrane bioreactor, and alum sludge can

increase the phosphorus removal efficiency [26, 27]. Membrane fouling is an unavoidable phenomenon that affects membrane performance during membrane separation operation, which refers to the adsorption and accumulation of pollutants on the membrane surface or in membrane pores, and the concentration polarization caused by it. The appearance of membrane fouling not only reduces the membrane flux, but also changes the membrane's ability to intercept and separate solutes [28, 29]. The dewatered alum sludge-based aerobic membrane reactor was designed for mustard tuber wastewater treatment in this study. By studying the influence of organic loading rate (OLR) on COD, $\text{NH}_4^+\text{-N}$ and soluble phosphate (SP), the treatment efficiency of this novel reactor for high salinity wastewater was evaluated.

Materials and methods

Experiment setting

The schematic of aerobic membrane reactor based on dehydrated alum sludge is shown in Figure 1. The effective volume of this reactor was 405 L, with size of $L \times B \times H = 1.08 \times 0.75 \times 0.50$ m. This reactor was made of Polyvinyl chlorideboard, which includes aerobic zone and membrane zone, and the length ratio of each zone was 7:2. Aerobic zone included porous aeration head and semi-soft biofilm-packing with 30 % biofilm density. Membrane zone included 2-3 mm aperture aeration pipe and polyvinylidene fluoride hollow fiber membrane. The membrane area of this hollow fiber membrane was 1 m^2 .

High-efficiency halotolerant bacteria in this aerobic membrane reactor based on dehydrated alum sludge were selected from the wastewater produced in the processing of pickling mustard tuber. Such bacteria would be inoculated into the aerobic zone of this aerobic membrane reactor.

Fig. 1. Schematic of aerobic membrane reactor based on dehydrated alum sludge: 1 - bucket, 2 - submersible pump, 3-6 - flowmeter, 7 - vacuum meter, 8 - biofilm-packing, 9 - fiber membrane, 10-11 - aeration pump, 12 - dump pump

Dewatered alum sludge and wastewater

The experimental dewatered alum sludge was collected from the industrial filter press of the sludge dewatering unit of a local water treatment plant in Chongqing, China, where aluminium sulphate was used as coagulant. Thereafter, the dewatered alum sludge was air-dried at room temperature for 10 days. The air-dried dewatered alum sludge was then ground and sieved to prepare the sludge for batch P-adsorption test, and the percentage of dewatered alum sludge added to biological sludge was 2-5 %. The main physicochemical properties of dewatered alum sludge were shown in Table 1. During the experimental process, the influent salinity mass fraction was controlled to be 4.0, 4.5, 5.0, 5.5, 6.0, 6.5 and 7.0 %, respectively. The acclimatization process under a specific salinity mass fraction was considered to be finished if the COD removal rate could reach 80.0 % stably. Then, the influent salinity mass fraction would be raised to the next level until it reached 7.0 %.

Table 1

Main physicochemical properties of dewatered alum sludge

Index	Total phosphorus (TP)	Active aluminum (Al _{ox})	Active iron (Fe _{ox})	Al	Fe	Ca	Mg
Concentration [g/kg]	0.48	56.65	8.56	58.70	9.25	5.36	5.45

The experimental wastewater was taken from the second pickle liquid of Fuling Mustard Tuber Group by Share Ltd, China; detailed composition was listed in Table 2. During the manufacture process of mustard tuber, the first pickle liquid contained more impurity, which could accelerate membrane fouling of experimental equipment. At the same time, the third pickle liquid possessed a relatively higher salinity, which could be reused in the production of mustard tuber sauce.

Table 2

The quality of experimental wastewater

Parameter	Concentration
COD [mg/L]	3300-3900
Total nitrogen (TN) [mg/L]	320-380
NH ₄ ⁺ -N [mg/L]	140-210
SP [mg/L]	50-62
Salinity [%]	6.8-7.3
Suspended solids (SS) [mg/L]	340-420

Experimental procedure

Parallel tests were carried out using three groups of reactors. All reactors in this experiment were operated in intermittent mode. The influent was transported into the reactor through submersible pump, and floating ball was used to control liquid level. The effluent of this reactor was transported through self-priming pump, and the working state of this pump was running for 8 min and stopping for 1 min. The effect of different trans-membrane pressure (TMP) on membrane fouling rate was studied in previous experiments, and the optimal TMP was selected in this experiment. Therefore, TMP in this experiment is a fixed value. The dissolved oxygen concentration of aerobic zone, aeration intensity (ratio of air to water) of membrane zone, TMP of fiber membrane and temperature were 4.0-5.0 mg/L, 12 m³/m²/h, 20 kPa and 30 °C, respectively. As tank volume and

influent concentration remained unchanged, the OLR depended on influent flow rate. According to preliminary test results and water quantity, the OLRs of this experiment were determined as 1.0, 2.0 and 3.0 kg COD/m³/day, and the influence of different OLR on the removal of COD, NH₄⁺-N and SP was studied in this study.

Analyses

Detection parameters during the experiment consisted of the following items: COD, NH₄⁺-N and SP. These parameters were analysed on the basis of standard methods for the Examination of Water and Wastewater which was released by American Public Health Association [30]. Samples were analysed with the standardised photometric cuvette tests from Hach-Lange for COD. For SP determination, sample pretreatment including filtration through 0.45 mm Millipore filters was necessary due to the large quantities of solids were present. Removal efficiencies were obtained by calculating the percentages of corresponding pollutant removal from the influent concentrations. In this study, the effluent concentration needs to meet Chinese “Wastewater quality standards for discharge to municipal sewers”, and the different level of Chinese “Wastewater quality standards for discharge to municipal sewers” is presented in Table 3.

Table 3
Quality of Chinese wastewater quality standards for discharge to municipal sewers

Parameter	Concentration		
	A-level	B-level	C-level
COD [mg/L]	500	500	300
NH ₄ ⁺ -N [mg/L]	45	45	25
Total phosphorus (TP) [mg/L]	8	8	5

Results and discussion

COD removal under different OLRs

The influent and effluent COD concentrations of dewatered alum sludge-based aerobic membrane reactor under different OLRs were shown in Figure 2.

It can be seen that when the influent COD concentration is 3480 mg/L, the COD removal efficiency shows a gradual decreasing trend with the gradual increase of OLR. The maximum removal efficiency was 96.6 % when the OLR was 1.0 kg COD/m³/day, and the minimum removal efficiency was 72.1 % when the OLR was 3.0 kg COD/m³/day. At the same time, when the OLR was more than 2.0 kg COD/m³/day, the COD concentration of effluent cannot meet the B-grade standard of Chinese “Wastewater quality standards for discharge to municipal sewers”. Considering all factors, the OLR of 2.0 kg COD/m³/day is the best.

With the increase of OLR, the number of microorganism increased, which lead to the increase of biofilm thickness. Meanwhile, this lead to anaerobic decomposition of biofilm's interior, which accelerated biofilm abscission [31]. At the same time, high OLR would result in uncompleted degradation due to poor biodegradability level [32]. Aloui et al. [33] adopted an activated sludge unit to treat an industrial wastewater containing 1-6 % salt, and investigated the influence of OLR on COD removal rate after 18 days of treatment in continuous mode. The COD removal rate decreased with the increase of OLR, and a total detoxification of the saline wastewater was achieved with salt concentrations below

2 % and OLR under 855 mg/L/day. Furthermore, the increase of OLR might also cause a partial disintegration of granules, thus resulting in the increase in effluent COD concentration [34-36].

Fig. 2. COD removal under different organic loading rate

$\text{NH}_4^+\text{-N}$ removal under different OLRs

The influent and effluent $\text{NH}_4^+\text{-N}$ concentrations of dewatered alum sludge-based aerobic membrane reactor under different OLRs were shown in Figure 3.

As can be seen from Figure 3, when the influent $\text{NH}_4^+\text{-N}$ concentration was 169 mg/L, the $\text{NH}_4^+\text{-N}$ removal efficiency showed a gradual decreasing trend with the gradual increase of OLR. The maximum removal efficiency was 94.1% when the OLR was 1.0 kg COD/m³/day, and the minimum removal efficiency was 74.0% when the OLR was 3.0 kg COD/m³/day. At the same time, even if the OLR was more than 2.0 kg COD/m³/day, the $\text{NH}_4^+\text{-N}$ concentration of effluent can still meet the B-grade standard of Chinese “Wastewater quality standards for discharge to municipal sewers”. Considering all the above factors, we believe that the OLR value of 3.0 kg COD/m³/day is relatively reasonable.

The analysis showed that, the higher OLR caused the more labile organic compounds content, which lead to the competition of heterotrophic bacteria and autotrophic bacteria, eventually caused the decrease of nitrification ability and the enhancement of denitrification ability [37]. Meanwhile, due to the dependence of nitrification on the supply of oxygen, the nitrification process of organic compounds would be inhibited when the OLR exceeded oxygen transfer rate [38, 39]. Jiang et al. [40] used food waste to study the influence of pH, temperature, and OLR on $\text{NH}_4^+\text{-N}$, and found that the concentration of $\text{NH}_4^+\text{-N}$ was lower due to a lower OLR and death of some fermentative microorganisms.

Fig. 3. $\text{NH}_4^+\text{-N}$ removal under different organic loading rate

SP removal under different OLRs

The influent and effluent SP concentrations of dewatered alum sludge-based aerobic membrane reactor under different OLRs were shown in Figure 4.

Fig. 4. SP removal under different organic loading rate

Figure 4 was a description of the decrease of SP under different organic loading rate. It can be seen that when the influent SP concentration was 56 mg/L, the removal efficiency of SP showed a gradual decrease trend with the gradual increase of OLR, and the maximum removal efficiency was 46.4 % when the OLR was 1.0 kg COD/m³/day. Even so, the SP concentration in wastewater cannot meet any level of Chinese “Wastewater quality standards for discharge to municipal sewers”.

In this experiment, no excessive sludge was discharged during the operation period, and the demand for phosphate of microorganism was small. Therefore, the removal of SP was dependent on phosphate reduction system rather than phosphate accumulating organisms [41]. At the same time, when OLR changed from 1.0 to 3.0 kg COD/m³/day, the effluent pH increased from 8.0 to 8.9, which would inhibit the activities of phosphate reduction bacteria [42]. However, the decreasing tendency of TP concentration in this experiment was not obvious. This may suggest that phosphate ions replaced -OH on the surface of dewatered alum sludge, which deduced that ligand exchange was the dominating pathway for phosphate removal [43, 44].

Conclusion

The dewatered alum sludge-based aerobic membrane reactor operating at 30 °C and 20 kPa of TMP was used to treat mustard tuber wastewater with salinity of 6.8-7.3 %. The effluent concentration of COD, NH₄⁺-N and SP all increased while OLR was changed from 1.0 to 3.0 kg COD/m³/day, and the effluent COD and NH₄⁺-N concentration except for SP could reach the B-level of Chinese “Wastewater quality standards for discharge to municipal sewers” when OLR was less than 3.0 kg COD/m³/day. At the same time, the change of temperature and pH in this experiment was very small, which has little effect on the removal of these experimental parameters. These data can effectively show that the aerobic membrane reactor based on dehydration alum sludge is a promising biological measure for treating high salinity wastewater.

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