International Journal of Chemical, Materials and Biomolecular Sciences
ISSN: 2415-6620
Vol:8, No:6, 2014

Nutrients Removal Control via an Intermittently Aerated Membrane Bioreactor

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Abstract**—**Nitrogen is among the main nutrients encouraging the growth of organic matter and algae which cause eutrophication in water bodies. Therefore, its removal from wastewater has become a worldwide emerging concern. In this research, an innovative Membrane Bioreactor (MBR) system named "moving bed membrane bioreactor (MBMBR)" was developed and investigated under intermittently-aerated mode for simultaneous removal of organic carbon and nitrogen.

Results indicated that the variation of the intermittently aerated duration did not have an apparent impact on COD and NH_4^+–N removal rate, yielding the effluent with average COD and NH_4^+–N removal efficiency of more than 92 and 91% respectively. However, in the intermittently aerated cycle of (continuously aeration/0s mix), (aeration 90s/mix 90s) and (aeration 90s/mix 180s); the average TN removal efficiency was 67.6%, 69.5% and 87.8% respectively. At the same time, their nitrite accumulation rate was 4.5%, 49.1% and 79.4% respectively. These results indicate that the intermittently aerated mode is an efficient way to controlling the nitrification to stop at nitrition; and also the length of anoxic duration is a key factor in improving TN removal.

Keywords**—**Membrane bioreactor (MBR), Moving bed biofilm reactor (MBBR), Nutrients removal, Simultaneous nitrification and denitrification.

I. Introduction

WATER pollution is a serious environmental and ecological problem because it may threat human health and cause eutrophication of aquatic plants. Organic matters and nitrogen are keys compounds that results in water pollution. Thus, the immediate and nuisance-free treatment for their removal has become the main target in recent environmental policy to protect public health and the environment.

To deal with the excess of pollutant contain in wastewater, there is a need to find adequate harmless elimination technologies with advantages of simple process, high efficiency and less energy consumption. The use of biological technologies has been reported in both pilot plant studies and full-scale plants; and has shown to be more effective, cost-effective, relatively inexpensive, and environmentally sound ways to control water pollution [1], [2]. Indeed, based on the microbial nitrogen cycle and the metabolism of inorganic nitrogen compounds, many biological technologies and processes have been developed and widely adopted for nutrients removal from wastewater [3], [4].

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The moving-bed-biofilm reactor (MBBR) is biological nutrients removal reactors designed to use carrier biofilm which offer the advantages of the biofilm process (compact, stable removal efficiency, simplicity of operation) without channeling and clogging of medium. Therefore, It has been found suitable to treat biologically various wastewaters [5], [6], such as dairy wastewater [7], [8]; and municipal wastewater [8].

Furthermore, membrane bioreactor (MBR), commonly known as the combination of membrane filtration and biological treatment using an activated sludge, have also been widely studied for wastewater treatment [9]. It has many advantages including small footprint and reactor requirements, high effluent quality, good disinfection capability, higher volumetric loading and less sludge production [10]. MBR has ability to operate at higher biomass concentrations and provide better retention of slow growing microorganisms [11].

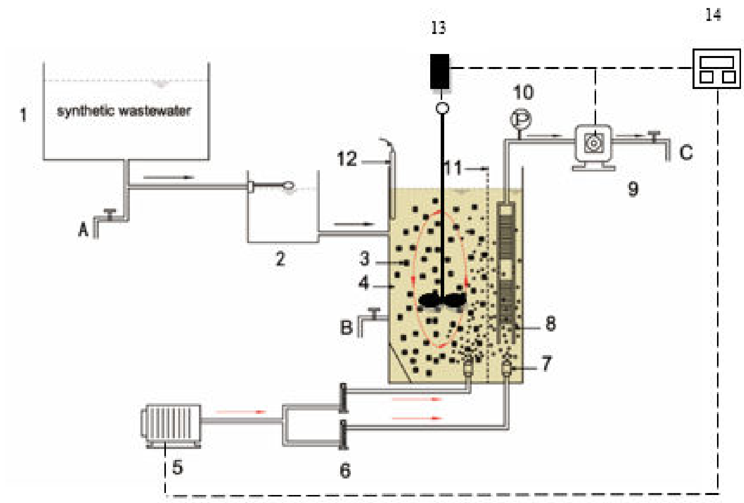
However, with the effluent discharge standards having become more stringent (e.g. <10mg total nitrogen/L), both conventional processes (MBBR and MBR) are far to meet the new requirements. Thus, as an alternative solution to improve the conventional features, this research developed a new biological nutrients removal technology by combining Moving Bed Bio Reactor (MBBR) and Membrane Bioreactor (MBR) features in one system called Moving Bed Membrane Bioreactor (MBMBR). It investigated its feasibility for simultaneous removal of residual chemical oxygen demand (COD) and nitrogen under various intermittently aerated operational schemes.

II. Materials and Methods

A. General Experimental Set-Up

Fig. 1 shows the schematic diagram of the experimental apparatus (MBMBR) used for this research. The reactor was made of Plexiglas with a working volume of 30 L.

A piece of clapboard with bores was fitted in the reactor, which divided the reactor into two parts with a volume ratio of 3:1. The membrane module was fixed inside the small part of the reactor for solid-liquid separation. The media carriers with internal walls was added (45% of the reactor volume) in the biggest part of the reactor in order to achieve a high inner square footage for microorganisms.



1. Feeding tank 2. Balance water box 3. Carrier elements 4. Activated sludge+ synthetic wastewater 5. Air pump 6. Air flow meter 7. Air diffuser 8. Membrane module 9. Water pump 10. Pressure gauge 11. Sieve 12. Electric heater 13. Agitator 14.Timer A. Influent sample collector B. Bioreactor sample collector C. Effluent sample collector

Fig. 1 Schematic of the moving bed membrane bioreactor (MBMBR)

Table I shows the specifications of the hollow fiber membranes and media-carriers used in this research.

TABLE I
SPECIFICATIONS OF THE HOLLOW FIBER MEMBRANE AND MEDIA-CARRIERS

Specifications of the Membrane	Membrane material	Polypropylene
	Type	Hollow fibers
	Pore size	0.1 µm
	Fiber length	170 Mm
	External diameter	400µm
	Wall thickness	40 µm
	Flux rate of pure water at 0.15Mpa, 25°C	100-120 $Lh^{-1}m^{-2}$
	Total membrane surface area	0.2 × 2 = 0.4 m^2
Specifications of the Media-Carrier	Material	Polyethylene
	Type	Hollow cylinder
	Diameter	20 mm
	Height	18 mm
	Specific Surface Area	900 m^2/m^3
	Density	0.2727g/cm^3

The feed water flowed into the MBMBR by gravitational force and its level was controlled at constant volume by the balance-box. A peristaltic pump, was used to pump permeate water from the reactor. An air pump was used to inject air to the reactor; and the alternation between aerobic and anaerobic conditions was created through the modulation of the air pump and the agitator intermittently controlled by a timer-controlled power supply system. A ceramic heater furnished with a thermostat device was used sometimes to maintain the desired temperature (especially during winter periods).

B. Operating Conditions

The synthetic wastewater fed to the reactor was made according to Table II.

The reactor was run for different medium compositions and the variations in operational parameters are summarized in Table III. The reactor was inoculated with activated sludge taken from the secondary settling tank of the municipal waste water treatment plant of Chun-liu, Dalian (about 3000mg/L ±150mg/L at the start-up of each operating run).Sometimes sodium bicarbonate (NaHCO3) was used as a buffer to adjust the mixed liquor pH between 7 and 8.

TABLE II
SYNTHETIC WASTEWATER CHARACTERISTIC

Compounds	(mg/L)	Compounds	(mg/L)
$CaCl_2$, $2H_2O$	22.0	$CuSO_4$, $5H_2O$	0.03
$MgSO_4$, $7H_2O$	25.0	KI	0.03
$FeSO_4$, $7H_2O$	20.0	$MnCl_2$, $4H_2O$	0.12
K_2HPO_4	22.0	$Zncl_2$	0.057
ETDA	20.0	Na_2MoO_4,$2H_2O$	0.06
$Fecl_3$, $6H_2O$	1.5	$CoCl_2$, $6H_2O$	0.15
H_3BO_3	0.15	$NaCl_2$	25.0

TABLE III
OPERATION PARAMETERS IN THE EXPERIMENT[a]

Parameters	Run 1	Run 2	Run 3
Number of operational days	1-20	21-40	41-127
$COD_{INFLUENT}$ (mg/L)	237.6 (21.2)	233.7 (32.3)	228.2 (28.5)
$TN_{INFLUENT}$(mg/L)	43.2 (10.2)	43.3 (9.4)	43.1 (7.5)
COD/TN	5.5	5.4	5.3
HRT (hours)	16	16	16
SRT (days)	15	15	15
pH	7.6–8.5	7.6–8.5	7.6–8.5
Mixed liquor temperature	25℃	25℃	25℃
Aeration rate	0.45 m^3/h	0.45 m^3/h	0.45 m^3/h
Suction type	Continuously	Continuously	Continuously
Cycle time (second)			
Aerobic	Continuously	90	90
Anaerobic	-	90	180

[a]Standard deviation is given between parentheses

At the end of each run, to provide similar condition and performance to the membrane module, the membrane modules are taken from the bioreactors and are cleaned by tap water and immersed in 0.3% NaClO solution for one day to get a permeability recovery more than 95%. Then the membrane modules are equipped in the reactor again for the next experimental phase.

C. Analytical Methods

Samples were collected from influent and effluent ports of the reactor, to analyze various parameters including mainly: chemical oxygen demand (COD), ammonia nitrogen (NH_4^+-N); nitrite nitrogen (NO_2^--N); nitrate nitrogen (NO_3^--N), total nitrogen (TN).

COD, NH_4^+-N, NO_2^--N, NO_3^--N and TN were analyzed according to standard methods for the analysis of water and wastewater [12]. Beside the sampling analysis some analysis due to reactor working set up were perform daily such as: pH, DO, permeate flux rate, TMP, water flux rate, temperature, etc. DO and pH in the reactor were measured by a DO meter (YSI 55/12 FT, USA) and a pH meter (Sartorius PB-10, Germany), respectively.

III. Results and Discussion

The long term operational period (127 days) of the reactor was to investigate the effect of aerobic duration/anoxic duration on COD, NH_4^+–N and TN removal efficiencies. Indeed, the principle of this reactor process is that small floating carrier elements provide a surface for microbial growth in a totally mixed reactor while the membrane ensure the liquid-suspended elements separation. The reactor is fed from the feed water tank which contains the synthetic wastewater and the water flows through the packed bed of solid grains (media carriers). Flowing through the packed bed, the wastewater undergoes biological treatment by contacting the aeration provided at the bottom of the reactor (Aeration also promote turbulence for sludge mixing and scours the membrane surface from fooling). The biological treated water can then be filtered from suspended solid through membrane filtration which works under water pump's action.

A. Biological Organic Substance Removal

Fig. 2 illustrates the variations of COD concentration and its removal efficiencies in the intermittently aerated MBMBR. Results showed good performance on organic carbon removal with COD removal efficiencies of 92.8% in average. The results also indicate that changes in aerobic duration did not show any significant influence on COD removal. Indeed, major part of COD was consumed during anaerobic phase, and microorganisms stored COD as PHB under anaerobic condition for later use during the aerobic phase. So, at the end of aerobic phase, the removal efficiency could reach 92.8% on average. It indicated that attached-growth bio-film and suspended growth sludge in the MBMBR have perfect ability of organic carbon removal.

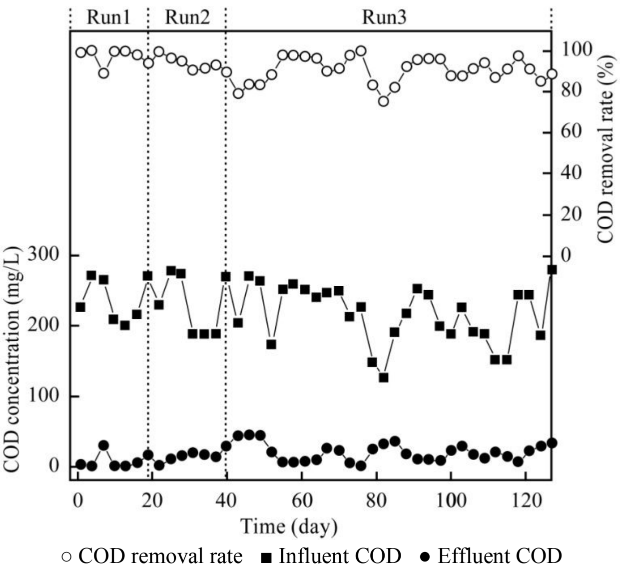


Fig. 2 COD concentrations and its removal efficiencies in intermittently aerated MBMBR

B. Biological Nitrogen Removal

Fig. 3 illustrates the profile of nitrogen concentrations as well as its removal efficiencies in the intermittently aerated MBMBR.

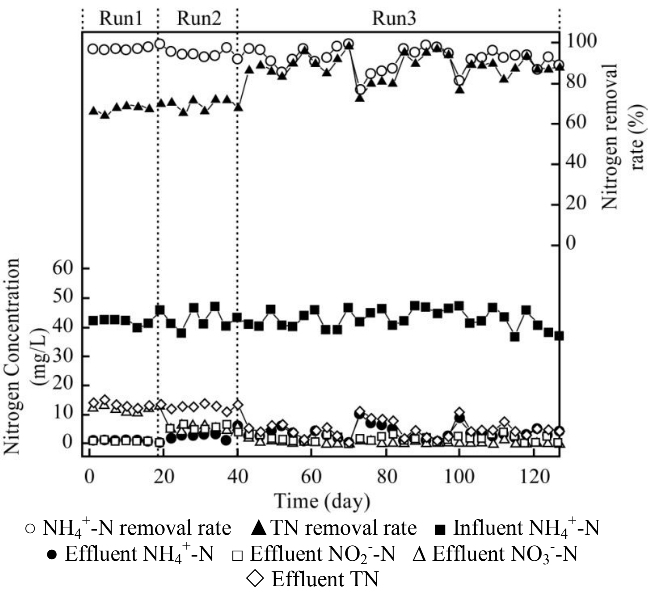


Fig. 3 Nitrogen concentrations and removal efficiencies in the intermittently aerated MBMBR

From Fig. 3, results indicated that:

– During run 1 (under continuously aerated mode), the effluent showed almost complete removal efficiency for NH_4^+–N averaging 97.0%, whereas the average TN removal efficiency was only 67.6%. The Nitrite accumulation rate (NAR) was only 4.5%. These results indicate that the nitrification was full-range nitrification and the main element was nitrate. In the nitrification step, Oxygen plays a role as the terminal electron acceptor for nitrifying bacteria. Indeed, under strict aerobic conditions, the ammonium is oxidized to nitrite and then to nitrate which use molecular oxygen as electron acceptor, ammonia or nitrite as energy source and carbon dioxide as Carbon source. As a result, there was full rang nitrification and the main product was nitrate.

- In run 2, the intermittently aerated mode (aeration 90s/mix 90s) was adopted. Results showed a slight decrease of NH_4^+–N removal efficiency and averaged 94.0%. The average TN removal efficiency raised to 69.5% and the NAR increased to 49.1%. Although the TN removal was not significantly improved, the nitrite accumulation was performed gradually under the intermittently aerated mode, which indicates that the intermittently aerated mode was an effective approach to controlling the rate of nitrite.
- With the adjustment of the intermittent time, Run 3 (aeration 90s/mix 180s), the NH_4^+–N removal efficiency averaging 91.8%, The NAR increased to 79.4% and the TN removal efficiency averaged 87.8%. These results indicate that increasing the mixing time is effective in improving TN removal efficiencies.
- In general, the change in anoxic duration did not have an obvious influence on NH_4^+–N removal. Indeed, most of the organic substances had been removed in anaerobic phase, hence, autotrophic bacteria could grow easily in aerobic phase, resulting in a good performance on NH_4^+-N removal. In the aerobic phase, the TN removal mainly depends on SND performance, which resulted from DO concentration gradients arising from diffusional limitations. Nitrification took place at the carrier interface, which was an aerobic layer, and denitrification occurred in the deeper layer of the biofilm, where anaerobic conditions were present. In the anaerobic phase, the residual nitrate and nitrite could be removed by using the influent COD as substrate. Therefore, in order to ensure simultaneous nitrogen, the SND performance must be enhanced during aerobic phase. In MBMBR, along with the attached-growth biofilm became thicker, the SND performance improved and the effluent TN concentration maintained at a low level.

IV. Conclusion

An intermittently aerated moving bed membrane bioreactor (MBMBR) was developed and investigated for simultaneously removing organic carbon and nitrogen in wastewater. Results demonstrated that intermittent aeration was an effective approach to achieve nitrition and the lengthening of anoxic duration is also a key factor affecting TN removal. The results suggest that the MBMBR system can successfully achieve stable and simultaneous removal of organic and nitrogen, however, the system is to be optimized for more effective nitrogen removal performance.

Acknowledgment

Financial supports from the National Natural Science Foundation of China (NSFC) (Grant no.40801228) and Ministry of higher Education and Scientific Research (Republic of Benin), are highly appreciated.

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