

Research Article

Effect of Gas/Water Ratio on the Performance of Combined Cylindrical Anoxic/Aerobic Moving Bed Biofilm Reactors for Biological Nutrients Removal from Domestic Wastewater by Fully Nitrification-Denitrification Processes

^{1,2}Husham T. Ibrahim, ¹HEQiang and ²Wisaam S. Al-Rekabi

¹Faculty of Urban Construction and Environmental Engineering, Chongqing University, Chongqing 400045, P.R. China

²Faculty of Engineering, Basra University, Basra, Iraq

Abstract: In this research the continuously up-flow pilot scale Moving Bed Biofilm Reactor (MBBR) which was consists of combined cylindrical anoxic/aerobic MBBR in nested form with anoxic/aerobic volume ratio equal to 0.16 under fully nitrification-denitrification process were used to treated 4 m³/day of domestic wastewater in Chongqing city at Southwest China. The treatment must be satisfactory to meet with grade B of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002). Both the anoxic and aerobic reactors were filled to 50% (v/v) with Kaldnes (K1) biofilm carriers to attach and retain biomass. The reactors was operated under fully nitrification-denitrification process without sludge returning into the system and only an internal recycling was performed from aerobic to anoxic reactor. After developing the biofilm on the carriers, the effect of gas/water ratio on biological nutrients removal from domestic wastewater was investigated by operated the reactors under 5 different gas/water ratio ranging from 5/1 to 24/1. During this operation mode, the favorite internal recycle ratio and Hydraulic Residence Time (HRT) to eliminate nitrogen compounds were 100% of inflow rate and 6.2 h, respectively. The experiment results showed that optimum value of the gas/water ratio for simultaneous organic carbon and nutrients removal was equal to 7/1. In this gas/water ratio, the average removal efficiencies were 92.67, 99.12, 71.37 and 90.49% for COD, NH₄⁺-N, TN and TP, respectively, while the average Dissolved Oxygen concentration (DO) in aerobic and anoxic MBBRs were 4.49 and 0.16 mg/L, respectively.

Keywords: Ammonium nitrogen, anoxic, attached biomass, autotrophic, biofilm, biological treatment, carrier, conventional activated sludge process, heterotrophic, nitrate, nitrite, nitrogen, phosphorus, sequencing batch moving bed biofilm reactor, suspended biomass

INTRODUCTION

The traditional methods for municipal wastewater were based on biological treatments (Kelly, 1996). Biological treatment of domestic wastewater by Conventional Activated Sludge process (CAS) has been practicing for more than 100 years. Whenever the discharge limit for various pollutants become more stringent (especially for nitrogen and phosphorus), the need to find a new treatment methods and process modifications to existing processes become more necessary. Even the activated sludge process modified numerous times in order to produce higher quality effluent the operational problems such as sludge bulking, sludge rising and Nocardia foam drastically reduced the efficiency of conventional activated sludge process. Higher hydraulic retention time requirement is another drawback of CAS and this leads to higher tank volumes, finally end up in large foot print.

Biofilm process was proved to be more reliable than suspended systems for organic carbon and nitrogen removal with no problems of suspended growth system. The most cost-effective nitrogen removal will probably be achieved by using rather compact biofilm processes (Anthonisen *et al.*, 1976; Helmer *et al.*, 1999). A biofilm process, which may be compact, is the one based on submerged biological filters. There are many reports concerning the possibility of biofilm process for treating wastewater, but the disadvantage of this system is the possibility of clogging of the biofilm media (Chen *et al.*, 1995; Al-Ghusain *et al.*, 1994; Halling and Jorgensen, 1993; Huang *et al.*, 1992; Rusten *et al.*, 1994, 1996, 2000).

The biofilm process with a highly specific surface and without the clogging problem is the moving bed biofilm reactor (Pastorlli *et al.*, 1999). In the last years, the Moving Bed Biofilm Reactor (MBBR) was introduced as an alternative and successful method to

Corresponding Author: Husham T. Ibrahim, Faculty of Urban Construction and Environmental Engineering, Chongqing University, Chongqing 400045, P.R. China

This work is licensed under a Creative Commons Attribution 4.0 International License (URL: <http://creativecommons.org/licenses/by/4.0/>).

treating different kinds of effluents under different conditions based on the biofilm principle that take advantage of both activated sludge process and conventional fixed film systems without their disadvantages (Andreottola *et al.*, 2002; Canziani *et al.*, 2006; Falletti and Conte, 2007). The first MBBR facility became operational in early 1990 in Norway and then this system developed in Europe and America. There are presently more than 400 large-scale wastewater treatment plants based on this process operation in 22 different countries all over the world (Maurer *et al.*, 2000). Reactor can be operated at very high load and the process is insensitive to load variations and other disturbances (Odegaard *et al.*, 1994; Delenfort and Thulin, 1997). Unlike most biofilm reactors, the reactor volume in the MBBR is totally mixed and consequently there is no dead or unused space in the reactor. In addition, this system has a small head loss and there is no need for recycling of biomass or sludge (Xiao *et al.*, 2007). The basic principle of the MBBR process is a continuous flow process which combine the two different processes (attached and suspended biomass) by adding biofilm small High Density Polyethylene (HDPE) carrier elements with a large surface area and a density slightly less or heavier than $1.0 \times 10^3 \text{ kg/m}^3$ into the tank for biofilm attachment and growth. The carrier media that is added for the growth of the attached can be freely moving inside the reactor because of the agitation setup by aeration (in aerobic reactor) or mechanical mixing (in anaerobic and anoxic reactor) (Odegaard *et al.*, 2000).

Wastewater containing high levels of phosphorus and nitrogen cause several problems, such as eutrophication, oxygen consumption and toxicity, when discharged into the environment. It is, therefore,

necessary to remove such substances from wastewaters in order to reduce their harm to the environment. Therefore the regulations on the Nitrogen (N) and Phosphorus (P) contents in wastewater discharge are increasingly more stringent, for controlling the rate of eutrophication in the aquatic environment. One of the important problems in modern wastewater treatment systems is ammonium removal. Biological Nutrients Removal (BNR) in both domestic and industrial wastewater treatments, generally seems increasingly necessary (Wang and Yang, 2004). Nitrogen compounds are usually removed from wastewater by a combination of two processes of nitrification and denitrification. In nitrification, ammonia is oxidized to nitrite and nitrate by two different groups of microorganisms. The first group of microorganisms, Ammonia Oxidizing Bacteria (AOB), converts ammonia to nitrite and after that, the second group, Nitrite Oxidizing Bacteria (NOB) oxidizes the intermediate product to nitrate. In the denitrification process, nitrate is first converted to nitrite (NO_2^-) and then to nitrous oxide or laughing gas (N_2O), Nitric Oxide (NO) and finally to nitrogen gas (N_2) (Wang and Yang, 2004). Usually, nitrite oxidation proceeds faster than ammonia oxidation, so that nitrite rarely increases in the environment (Rittmann and McCarty, 2001).

The aim of the present study was to evaluate the influence of gas/water ratio on the performance of a continuous up-flow pilot scale combined cylindrical anoxic/aerobic MBBRs for biological nutrients removal from domestic wastewater by fully nitrification-denitrification processes. The innovation in this technology was the construction of the reactors in the form of overlapping cylinders by placed the anoxic reactor inside the aerobic reactor with anoxic/aerobic

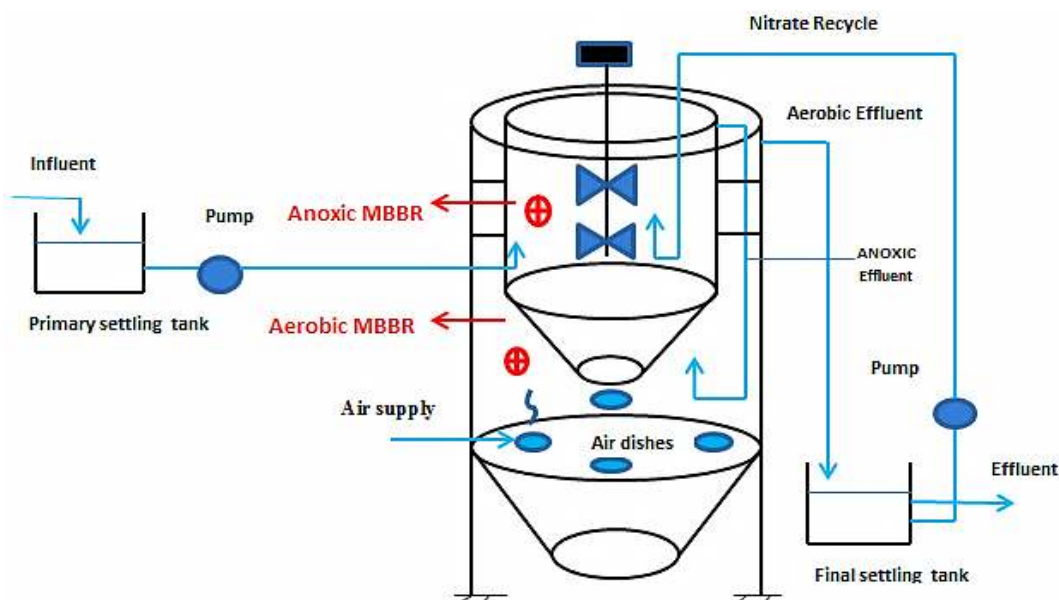


Fig. 1: Schematic diagram of pilot-scale MBBR system

volume ratio equal to 0.16; this led to reduce the required space for the establishment of the reactors.

MATERIALS AND METHODS

Experimental set-up: The experiments were conducted using two steel pilot scale MBBRs in nested form, including square primary settling tank (made of PVC 1×1×1 m), an anoxic reactor (R1) (D = 0.6 m and H = 0.9 m), an aerobic reactor (R2) (D = 1.2 m and H = 2 m) followed by a square final clarifier (made of PVC 1×1×1 m). No sludge recycling was implemented. The anoxic MBBR (R1) was used to achieve the denitrification processes by provide the major portion of nitrate removal, while the aerobic MBBR (R2) was built to provide nitrification. A sketch of the pilot-scale moving bed biofilm reactors is shown in Fig. 1 and some key parameters are listed in Table 1. Both anoxic and aerobic MBBRs were operated in an up-flow mode, Kaldnes (K1) media was used as a carrier in both reactors at a media fill ratio equal to 50%. The Kaldnes (K1) carrier elements are made of polyethylene (density 0.93 g/cm³) and shaped like small cylinders (about 25 mm in diameter and 10 mm long) with a cross inside to provide sites for active bacteria attachment in a suspended growth medium. The effective specific growth area is 500 m²/m³ at 100% filling grade, Table 2 show the characteristics of the Kaldnes (k1) media which used in this study. The biofilm carrier elements are kept suspended in the water by air from the diffusers in aerobic reactor and by means of propeller mixer in anoxic reactor. The propeller mixer consist of central, 2-blade double stirrer of 25 cm diameter and with blades placed at 20 and 40 cm below top-water level, the stirrer speed was 100 rpm. The carrier elements are retained by means of small sized sieve (about 2 mm opening). Aeration system consist of 4 fine bubble membrane diffuser (4 aeration dishes 220 mm in diameter) distributed equally on the perimeter of the reactor which are fixed at the height 0.3 m from the bottom of the reactor. Aeration was achieved by using air compressor model ACO-818 (the largest aeration capacity of 300 L/min) connected with the main air distribution pipe (UPVC pipe 25 mm in diameter) which connected each aeration dish with the other. The airflow to the reactor was measured by gas rot meter (model LZB-10WB 5~45 L/min) and regulated with a manual valve. Sampling ports were provided in each reactor by using DN10 UPVC pipes for sample collection in the top, middle and bottom. The reactors were built in the Dadukou wastewater treatment plant which is located in Dadukou district in Chongqing city at Southwest China. The domestic waste water reached to the primary settling tank from the preliminary treatment part in Dadukou wastewater treatment plant

Table 1: Technical data and key parameters for the anoxic-aerobic MBBRs

Parameter	Anoxic MBBR (R ₁)	Aerobic MBBR (R ₂)
Effective volume (m ³)	0.141	0.89
Filling ratio with bio-carriers (%)	50.000	50.00
Specific biofilm surface area (m ² /m ³)	250.000	250.00
Total biofilm surface area (m ²)	35.250	222.50
Flow rate (m ³ /day)	4.000	4.00
Flow direction	Up-flow	Up-flow

Table 2: The characteristics of the kaldnes (k1) media

Parameter	Value
Dimension (mm)	25×10
Surface area (m ² /m ³)	500
Filling ratio (%)	15-65
Density (g/cm ³)	0.93
Number/m ³	150,000
Voidage (%)	95
Oxidation efficiency of BOD ₅ (g BOD ₅ /m ³ .day)	6000
Hanging coefficient (g/carrier)	1.30

which is responsible for removal of wastewater constituents such as rags, sticks, floatable, grit and grease that may cause operational problems with the treatment operations. The main purpose of the establishment of the primary settling tank is to remove part of suspended matter and organic material by settling, this method protect the pumps and pipes from clogging also improve subsequent biological treatment and keep stable water quality. The anoxic MBBR was continuously received the domestic wastewater from the primary settling tank in the start-up mode and from both the primary settling tank and the final clarifier in steady state mode by using magnetic circulation pumps model MP-30RZM with maximum capacity and maximum head equal to 15-17 (L/min) and 8-11 (m) respectively. Both influent and effluent system pipes are made of Un-plasticized Polyvinyl Chloride (UPVC) with diameter equal to 20 mm (DN20). The effluent system of the anoxic MBBR consists of 5 (DN20) UPVC pipes which carry the water to the aerobic MBBR by gravity. The influent and recycle flow was measured by a glass flow meter model LZB-15 (25-250 L/h) for influent flow and glass flow meter model LZB-25 (40-400 L/h) for recycle flow and can be regulated by controlling the manual valves (DN20).

Operating procedure: The study was carried out using raw domestic wastewater supplied from the preliminary treatment part in Dadukou wastewater treatment plant. The quality of wastewater resulting from the various daily uses in Dadukou district at Chongqing city in China are given in Table 3. Seeding sludge was obtained from Dadukou municipal wastewater treatment plant. Firstly the collected sludge was screened with a sieve to remove coarse and inorganic particles, then sludge was aerated for 2 days at room temperature. After that the sludge was mixed with

Table 3: Characteristics of the domestic wastewater from Dadukou district at Chongqing city in China

Parameter	COD (mg/L)	NH ₄ ⁺ -N (mg/L)	TN (mg/L)	TP (mg/L)	pH
Value	76.5-430	24.31-70.8	28-74.5	1.88-8.27	6.8-7.58

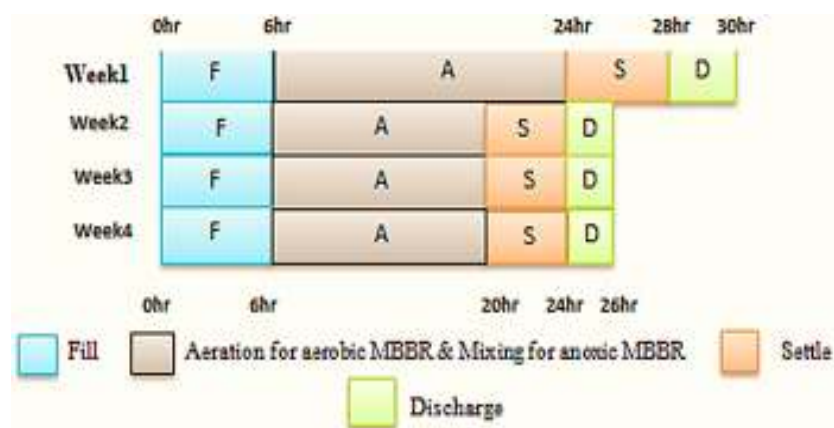


Fig. 2: Batch running cycle mode under start-up phase

wastewater by the ratio of 2/3 then filled 1/3 of effective volume for reactors by the mixed liquid. By this way the reactors are ready to batch operation mode for 4 weeks. The batch operation was used as start-up for growth of biofilm on carrier elements. After this period, the biofilm appeared on packing elements and MBBRs seemed to be ready for continuous operation. During the batch operation mode the reactors was operated as a Sequencing Batch Moving Bed Biofilm Reactor (SBMBBR) with fill period approximately equal to 6 h for all operation cyclic modes and gas/water ratio equal to 7/1. During the 1st week of operation mode the Mixed Liquor (ML) was continuously aerated in aerobic MBBR and mixing in anoxic MBBR for 18 h and then settled for 4 h after that water discharge with drainage ratio of 100% for 2 h, while during the 2nd, 3rd and 4th week the Mixed Liquor (ML) was continuously aerated in aerobic MBBR and mixing in anoxic MBBR for 14 h and then settled for 4 h after that water discharge with drainage ratio of 100% for 2 h. Figure 2 show the batch daily running cycle during start-up phase. At the end of 4th week the pilot plant was operated under continuous operation mode at Hydraulic Retention Time (HRT) of 6.2 h, with nitrate recycle ratio equal to 100% and gas/water ratio equal to 7/1, getting prepared for main start-up. At the end of 5th week the pilot plant was operated under continuous operation mode at 5 different gas/water ratio (5/1, 7/1, 10/1, 14/1 and 24/1) with Hydraulic Retention Time (HRT) equal to 6.2 h and nitrate recycle ratio equal to 100%. During this operation mode the temperature average values in both anoxic and aerobic MBBRs were 30.40 and 30.80°C respectively, the average value of the total Mixed Liquor Suspended Solids concentration (MLSS Total) in both anoxic and aerobic reactors

equal to 2855 and 3001.7 mg/L respectively, while PH average values were 7.54 and 7.44, respectively.

Sampling and analysis: Samples were collected from influent and effluent of MBBRs. The analytical techniques used in this study were performed according to the standard methods described in (State Environmental Protection Administration, 2002). Temperature, Dissolved Oxygen (DO) and PH were measured in each reactor by used Multi parameter Meter (HACH sension TM156). The Dissolved Oxygen (DO) was tested three times every day in both anoxic and aerobic MBBRs, in the anoxic MBBR the DO was tested in the top, middle and the bottom of the reactor then the average value was used, while in the aerobic MBBR the DO was tested in four points at middle of reactor according to the locations of the aeration dishes then the average value was used. Both the pH and the temperature was tested three times every day in both anoxic and aerobic MBBRs and the tests was done in the middle of the reactors. The samples of COD, ammonium nitrogen (NH₄⁺-N), Total Nitrogen (TN) and Total Phosphorous (TP) were measured on alternate days by used HACH DR5000UV Spectrophotometer. The assessment of the Total Suspended Solids concentration (TSS) on the fixed biomass elements was performed as follows: the attached biomass was removed from the 10 bio-carriers by putting them in a flask with demineralized water that was placed in an ultrasound bath for 45 min. After that the bio-carriers were rinsed with demineralized water and then the mixed liquid was filtered through 0.45 μm fiber filter and dried at 105°C and weighed. Because of the variability of carrier's dimension, the obtained value was referred to the total measured surface of the 10 bio-

carriers. TSS was assessed through the total surface in one cubic meter of reactor (Andreottola *et al.*, 2000a, b; Jahren *et al.*, 2002; Helness, 2007).

RESULTS AND DISCUSSION

In this research an experimental study to evaluate the application of fully nitrification/denitrification process in the continuous up-flow combined cylindrical anoxic/aerobic MBBRs system for the organic carbon and biological nutrients removal from domestic wastewater in Chongqing city at Southwest China. The

treatment must be satisfactory to meet with grade B of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) which show in Table 4. Operation and performance data are presented in Table 5 to 7 and shown in Fig. 3 to 12.

Rusten *et al.* (1995) reported that the degradation of organic matter at low DO concentrations in aerobic MBBR will slow down or stop the nitrification process. Heterotrophs and nitrifiers will compete for available oxygen and the rapidly growing heterotrophs will dilute (or wash out) the nitrifiers in the biofilm. The oxygen

Table 4: Discharge standard of pollutants for municipal wastewater treatment plant (GB/T18918-2002)

Parameter	COD (mg/L)	BOD ₅ (mg/L)	NH ₄ ⁺ -N (mg/L)	TN (mg/L)	PO ₄ ³⁻ -P (mg/L)
Grade A	50	10	5	15	0.5
Grade B	60	20	8	20	1

Table 5: Reactor performance in COD, NH₄⁺-N, TN and TP removal at different gas/water ratio in steady state operation

Gas/water ratio	COD			NH ₄ -N			TN			TP		
	INF. (mg/L)	EFF. (mg/L)	R. (%)	INF. (mg/L)	EFF. (mg/L)	R. (%)	INF. (mg/L)	EFF. (mg/L)	R. (%)	INF. (mg/L)	EFF. (mg/L)	R. (%)
5	361.50	23.50	93.50	48.90	4.90	89.98	54.03	31.33	42.01	2.49	0.63	74.51
5	311.50	28.50	90.85	62.31	7.20	88.44	68.53	30.74	55.14	3.83	0.94	75.50
5	145.50	24.60	83.09	25.61	2.01	92.15	28.00	17.19	38.62	6.72	1.36	79.76
5	280.50	31.00	88.95	47.78	5.70	88.07	53.60	31.95	40.39	3.70	0.97	73.80
7	347.50	15.30	95.60	59.50	0.40	99.33	65.70	15.40	76.56	2.83	0.48	83.05
7	178.60	20.80	88.35	36.80	0.40	98.91	37.58	13.74	63.44	3.37	0.39	88.42
7	318.50	16.90	94.69	58.80	0.20	99.66	62.40	16.30	73.88	4.35	0.31	92.87
7	244.10	19.40	92.05	38.21	0.54	98.59	43.76	14.60	66.63	7.52	0.18	97.61
10	240.00	16.40	93.17	57.34	3.30	94.24	58.20	30.92	46.87	4.62	0.85	81.58
10	330.00	11.40	96.55	44.30	2.30	94.81	51.30	31.61	38.38	1.88	0.77	59.04
10	368.00	9.20	97.50	34.87	2.80	91.97	35.13	22.61	35.64	2.32	0.92	60.33
10	161.50	23.50	85.45	53.04	4.10	92.27	74.50	39.21	47.37	6.70	0.74	88.95
14	315.00	7.50	97.62	53.10	5.40	89.83	57.40	43.60	24.04	2.90	1.10	62.07
14	291.50	8.70	97.02	59.20	6.10	89.70	64.70	35.97	44.40	3.60	1.12	68.89
14	222.50	16.20	92.72	28.30	1.30	95.41	35.60	27.69	22.22	5.70	1.69	70.35
24	377.50	4.50	98.81	46.09	5.80	87.42	48.94	35.62	27.22	5.82	1.85	68.21
24	255.00	9.10	96.43	42.64	4.20	90.15	45.60	35.98	21.09	3.86	1.53	60.34
24	190.00	12.70	93.32	54.90	7.20	86.89	62.10	44.17	28.87	2.41	0.99	58.91

INF.: Total influent; EFF.: Total effluent; R.: Total removal efficiency

Table 6: Average reactor performance in COD, NH₄⁺-N, TN and TP removal at different gas/water ratio in steady state operation

Gas/water ratio	COD			NH ₄ -N			TN			TP		
	Av. INF. (mg/L)	Av. EFF. (mg/L)	Av. R. (%)	Av. INF. (mg/L)	Av. EFF. (mg/L)	Av. R. (%)	Av. INF. (mg/L)	Av. EFF. (mg/L)	Av. R. (%)	Av. INF. (mg/L)	Av. EFF. (mg/L)	Av. R. (%)
5	274.75	26.90	89.10	46.15	4.95	89.66	51.04	27.80	44.04	4.19	0.98	75.89
7	272.18	18.10	92.67	48.33	0.39	99.12	52.36	15.01	70.13	4.52	0.34	90.49
10	274.88	15.13	93.17	47.39	3.13	93.32	54.78	31.09	42.07	3.88	0.82	72.48
14	276.33	10.80	95.78	46.87	4.27	91.64	52.57	35.75	30.22	4.07	1.30	67.10
24	274.17	8.77	96.19	47.88	5.73	88.78	52.21	38.59	25.73	4.03	1.46	62.49

Av.: Average value; INF.: Total influent; EFF.: Total effluent; R.: Total removal efficiency

Table 7: Standard deviation of reactor performance in COD, NH₄⁺-N, TN and TP removal at different gas/water ratio in steady state operation

Gas/water ratio	COD			NH ₄ -N			TN			TP		
	INF. S.D. (mg/L)	EFF. S.D. (mg/L)	R. S.D. (%)	INF. S.D. (mg/L)	EFF. S.D. (mg/L)	R. S.D. (%)	INF. S.D. (mg/L)	EFF. S.D. (mg/L)	R. S.D. (%)	INF. S.D. (mg/L)	EFF. S.D. (mg/L)	R. S.D. (%)
5	92.40	3.47	4.42	15.20	2.18	1.85	16.85	7.09	7.53	1.79	0.30	2.67
7	76.08	2.47	3.25	12.51	0.14	0.47	13.80	1.10	6.12	2.10	0.13	6.22
10	92.70	6.34	5.47	9.95	0.77	1.41	16.32	6.79	5.95	2.23	0.08	15.08
14	48.08	4.71	2.67	16.37	2.59	3.26	15.14	7.96	12.32	1.46	0.34	4.42
24	95.21	4.11	2.75	6.32	1.50	1.75	8.72	4.84	4.10	1.71	0.43	5.01

S.D.: Standard deviation; INF.: Total influent; EFF.: Total effluent; R.: Total removal efficiency

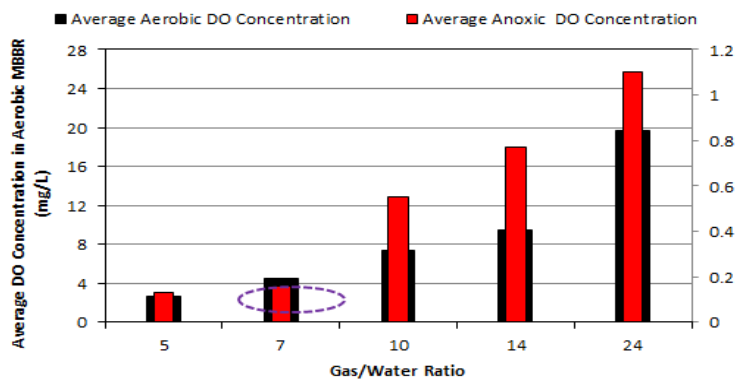


Fig. 3: Profile of average DO concentrations in aerobic and anoxic MBBRs versus gas/water ratio

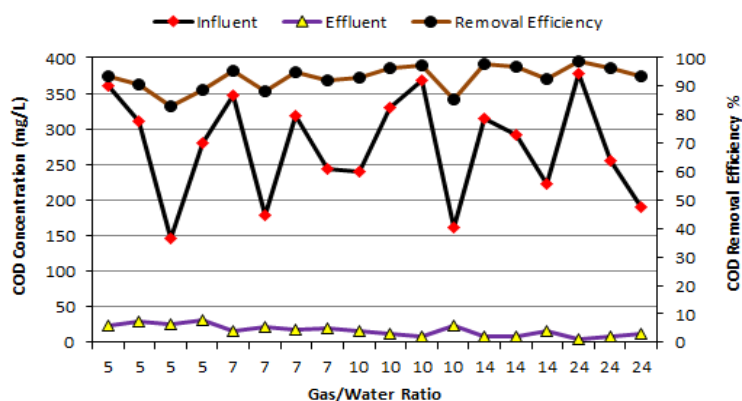


Fig. 4: Profile of COD concentration and removal efficiency variations versus gas/water ratio

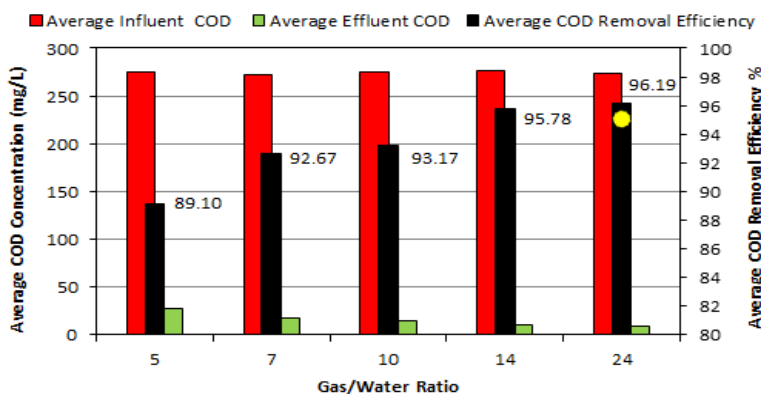


Fig. 5: Profile of average COD concentration and average removal efficiency variations versus gas/water ratio

concentration is mentioned as a very important limiting factor and could be used as a tool for nitrification (Ruiz *et al.*, 2003; Bernet *et al.*, 2001; Bae *et al.*, 2001). Low DO concentrations can affect the specific growth rate of both Ammonium Oxidizing Bacteria (AOB) and Nitrite-Oxidising Bacteria (NOB), depending on its oxygen saturation constant; however, its influence on the NOB, (1.1 mgO₂/L) is significantly greater than on the AOB (0.3 mgO₂/L) (Wiesmann, 1994). At low oxygen concentrations, changes in the population structure were observed by Park and Noguera (2004),

which could affect nitrite accumulation rates. Oxygen level for nitrite accumulation (Partially nitrification processes) is in the range of 0.5-1.5 mg O₂/L, while for nitrate accumulation (Fully nitrification processes) is over 2 mg/L (Ciudad *et al.*, 2005; Botrous *et al.*, 2004; Bernet *et al.*, 2001). Otherwise, the transport mechanisms in immobilized systems might even enhance then nitrite accumulation, since the oxygen is normally consumed in the first 50-100 μm of biofilms due to the deficient oxygen transfer into biofilms (Okabe and Watanabe, 2000) and an outward diffusion

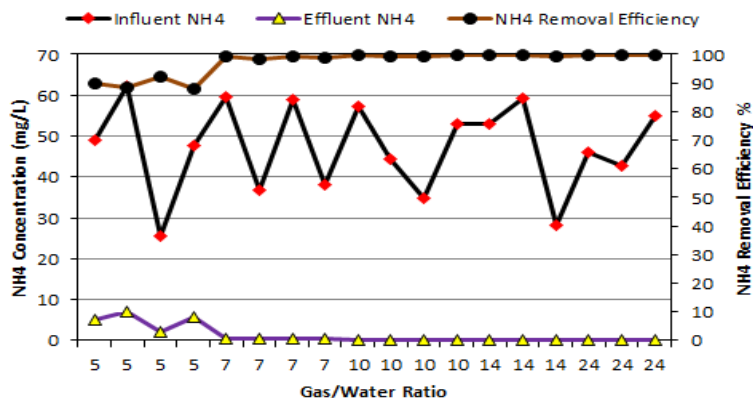


Fig. 6: Profile of NH_4^+ -N concentration and removal efficiency variations versus gas/water ratio

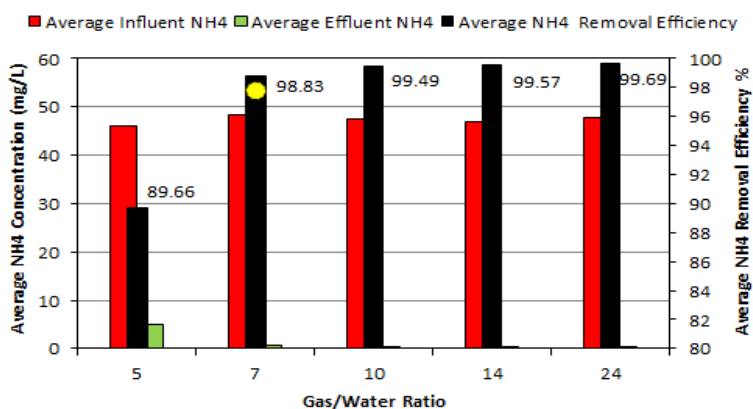


Fig. 7: Profile of average NH_4^+ -N concentration and average removal efficiency variations versus gas/water ratio

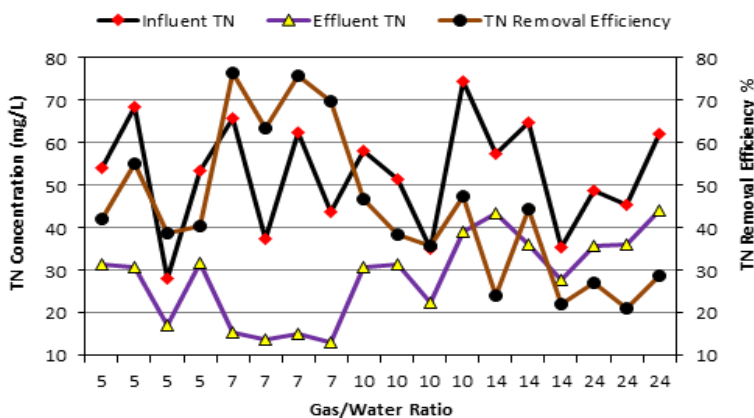


Fig. 8: Profile of TN concentration and removal efficiency variations versus gas/water

of the accumulated nitrite from the inside of the biofilm to the liquid bulk also occurs. Therefore, biofilm reactors with low mass transfer coefficient at the inter phase biofilm/liquid, such as the Rotating Disk Reactor (RDR) or biological aerobic filter (Lindemann and Wiesmann, 2000) and Moving Bed Biofilm Reactors (MBBRs).

The anoxic MBBR was designed to achieve the denitrification processes, while the aerobic MBBR was designed to achieve the nitrification processes. The

denitrification processes is very important process in the nitrogen removal by utilizing nitrite and nitrate as electron acceptors, DO can inhibit the denitrification reaction because oxygen functions as the electron acceptor for microorganisms over nitrate and aerobic conditions repress enzymes involved in denitrification (Zumft, 1997). Although high DO concentrations in the biofilm reactor are necessary to enhance the activity of nitrifying bacteria, denitrification is inhibited by oxygen (Hagedorn-Olsen *et al.*, 1994;

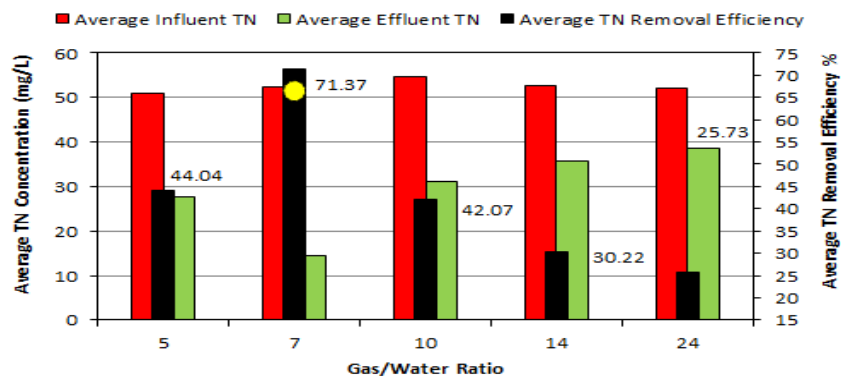


Fig. 9: Profile of average TN concentration and average removal efficiency variations versus gas/water ratio

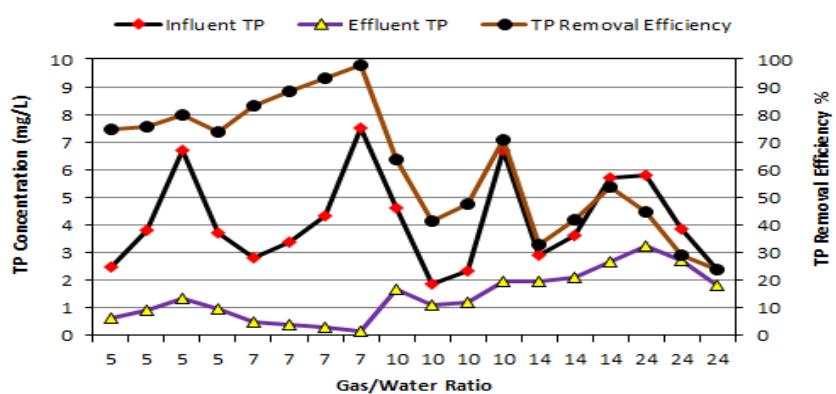


Fig. 10: Profile of TP concentration and removal efficiency variations versus gas/water ratio

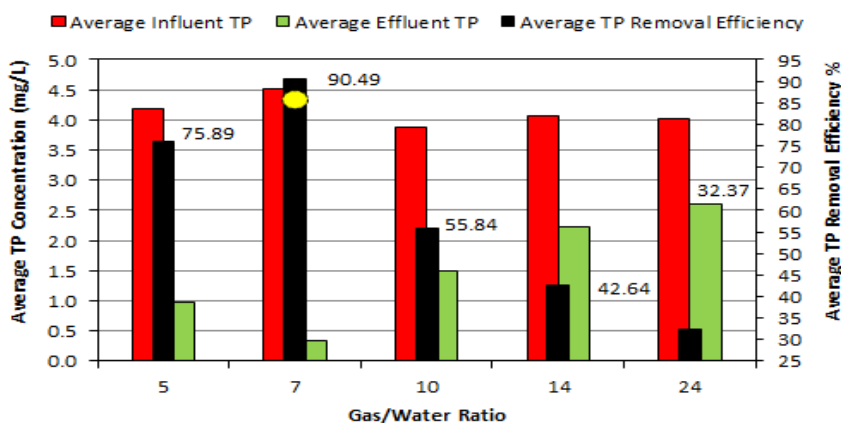


Fig. 11: Profile of average TP concentration and average removal efficiency variations versus gas/water ratio

Lie and Welander, 1994). When nitrification rate in aerobic MBBR increases more nitrate enters the anoxic MBBR and as a result more denitrification and subsequently more COD removal is achieved. Because the low DO level in anoxic MBBR, ammonia-oxidizers use nitrite as an artificial electron acceptor and generate nitrous oxide (N₂O) gas (Ritchie and Nicholas, 1972). Nitric Oxide (NO) gas is similarly produced by ammonia-oxidizers, but this activity is less sensitive to DO (Anderson and Levine, 1986). In the anoxic MBBR

some of the phosphate was removed by Denitrifying Phosphate-Accumulating bacteria (DNPAO) which using nitrate as electron acceptor and consumed some of the biodegradable organic matter, there must also be sufficient ammonium available for phosphate denitrification. By this way the anoxic MBBR consumed a part of the biodegradable Organic matter (COD) in order to removed nitrate and phosphate. Thus, reducing the pollutants loading rate in the aerobic MBBR and increase overall efficiency of the treatment.

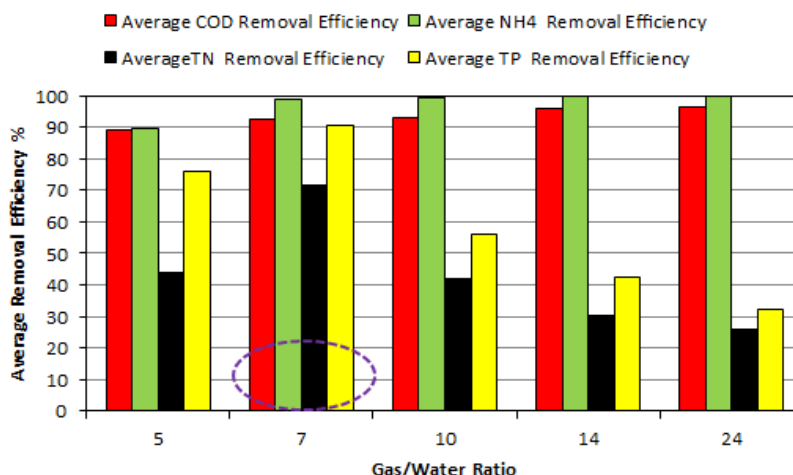


Fig. 12: Average total removal efficiency of COD, $\text{NH}_4^+\text{-N}$, TN and TP versus HRT

In the aerobic MBBR the dissolved oxygen was consumed by a competition between heterotrophic (COD removal), autotrophic (nitrification) and Phosphate-Accumulating Organism (PAO), while the biodegradable Organic matter (COD) was consumed by a competition between heterotrophic and Phosphate-Accumulating Organism (PAO). Figure 3 illustrated the DO concentrations in both aerobic and anoxic MBBRs under different Gas/water ratio. The amount of DO fed to the anoxic reactor due to the internal recycle flow from the aerobic reactor was influenced because DO in anoxic reactor was found increased when DO in aerobic reactor increased. As Gas/water ratio was increased from 5/1 to 7/1, the average DO concentrations in aerobic MBBR increased from 2.67 mg/L (S.D. = 0.21) to 4.49 mg/L (S.D. = 0.16), while the average DO concentration in anoxic MBBR increased from 0.13 mg/L (S.D. = 0.01) to 0.16 mg/L (S.D. = 0.02). When the Gas/water ratio increased from 10/1 to 24/1, the average DO concentrations in aerobic MBBR increased from 7.43 mg/L (S.D. = 0.35) to 19.68 mg/L (S.D. = 2.73), thus increases the average DO concentration in anoxic MBBR in the range from 0.55 mg/L (S.D. = 0.07) to 1.1 mg/L (S.D. = 0.13), makes the behavior of the anoxic MBBR like to be similar to that of the aerobic MBBR.

Organic Carbon (COD) removal: Total COD concentrations of influent, effluent and total removal efficiency versus Gas/water ratio are shown in Fig. 4 and Table 5, while the average performance of the MBBRs in COD removal is shown in Fig. 5 and Table 6 and 7. The daily concentration of COD in the feed were ranging from 145.5 to 377.5 mg/L (average = 274.37 and S.D. = 73.38) while the total effluent COD concentrations steadily decreased to the range 4.5 to 31 mg/L (average = 16.62 and S.D. = 7.59). As Gas/water ratio was increased from 5/1 to 24/1, the average total effluent COD concentrations decreased from 26.9 mg/L (S.D. = 3.47) to 8.77 mg/L

(S.D. 4.11), while the average total removal efficiency increased from 89.1% (S.D. = 4.42) to 96.16% (S.D. = 2.75). The results illustrated that the Gas/water ratio in range 5/1 to 24/1 did not significantly affect COD removal efficiencies and the total effluent COD concentration could meet with grade A of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) (50 mg/L).

Ammonium ($\text{NH}_4^+\text{-N}$) removal: Total ammonium concentrations of influent, effluent and total removal efficiency versus Gas/water ratio are shown in Fig. 6 and Table 5, while the average performance of the MBBRs in ammonium removal are shown in Fig. 7 and Table 6 and 7. It seems that the daily fluctuations of $\text{NH}_4^+\text{-N}$ concentration in the feed were so high while the total effluent $\text{NH}_4^+\text{-N}$ concentrations decreased to the range 2.01 to 5.7 mg/L at Gas/water ratio of 5/1, 0.47-0.6 mg/L at Gas/water ratio of 7/1, 0.17-0.27 mg/L at Gas/water ratio of 10/1, 0.11-0.23 mg/L at Gas/water ratio of 14/1 and 0.09-0.15 mg/L Gas/water ratio of 24/1. As Gas/water ratio was increased from 5/1 to 7/1, the average total effluent $\text{NH}_4^+\text{-N}$ concentrations decreased from 4.95 mg/L (S.D. = 2.18) to 0.53 mg/L (S.D. = 0.06), while the average total removal efficiency increased from 89.66% (S.D. = 1.85) to 98.83% (S.D. = 0.42). When the Gas/water ratio was increased from 10/1 to 24/1 the treated system became consist of two aerobic reactors due to increases in the average DO concentration in anoxic MBBR in range 0.55 mg/L (S.D. = 0.07) to 1.1 mg/L (S.D. = 0.13), the average total removal efficiencies were very close to 99%, while the average total effluent ammonium concentration are less than 0.25 mg/L, this value could comply with grade A of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) (5 mg/L). Finally the Gas/water ratio in range 5/1 to 24/1 did not significantly affect $\text{NH}_4^+\text{-N}$ removal efficiencies.

Total Nitrogen (TN) removal: The profile of TN concentration and removal efficiency variations versus HRT are shown in Fig. 8 and Table 5, while the average performance of the MBBRs in TN removal are shown in Fig. 9 and Table 6 and 7. As illustrated in Fig. 8 and 9 and Table 5 and 6, the Gas/water ratio in range 5/1 to 24/1 significantly affect TN removal efficiencies. As Gas/water ratio was increased from 5/1 to 7/1, the average total effluent TN concentrations decreased from 27.8 mg/L (S.D. = 7.09) to 14.38 mg/L (S.D. = 1.03), while the average total removal efficiency increased from 44.04% (S.D. = 7.53) to 71.37% (S.D. = 6.12). As Gas/water ratio was increased from 10/1 to 24/1, the average total effluent TN concentrations increased from 31.09 mg/L (S.D. = 6.79) to 38.59 mg/L (S.D. = 4.84), while the average total removal efficiency decreased from 42.07% (S.D. = 5.95) to 25.73% (S.D. = 4.1) because the high DO level in anoxic MBBR was suppress the denitrification process. Only the average total effluent TN at Gas/water ratio of 7/1 could meet with grade B of discharge standard of pollutants for municipal waste water treatment plant in China (GB/T18918-2002) (20 mg/L).

Total Phosphorus (TP) removal: The results of TP removal are shown in Fig. 10 and 11 and Table 5 to 7. The results indicate that the average overall phosphorus removal efficiency were increased from 75.89% (S.D. = 2.67) to 90.49% (S.D. = 6.22) as Gas/water ratio increased from 5/1 to 7/1. The anoxic/aerobic MBBRs show acceptable efficiency of TP removal from waste water only at Gas/water ratio of 5/1 and 7/1. At Gas/water ratio of 5/1 the average total effluent TP could meet with grade B of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) (1 mg/L), while at Gas/water ratio of 7/1 the average total effluent TP could meet with grade A of this standard (0.5 mg/L). According to the results, the Gas/water ratio significantly affect TP removal efficiencies only in range 10/1 to 24/1. Average total removal efficiency of COD, $\text{NH}_4^+\text{-N}$, TN and TP versus HRT are shown in Fig. 12.

CONCLUSION

From different tests in pilot-scale plants, the following experiences have been obtained with anoxic/aerobic MBBR:

- Major advantages of MBBR as compared to other systems are simplicity in operation, low space requirement, stability, reliability, good settle ability, low head loss, no bulking and lack of backwash requirement.

- The reactor has demonstrated its capability for nitrification, denitrification and organic removal process of a broad range of ammonia and COD.
- The average total effluent of TN concentrations could not meet with grade B of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) at Gas/water ratio of 5/1, 10/1, 14/1 and 24/1, while could meet with grade A of this standard at Gas/water ratio of 7/1.
- The average total effluent of TP concentrations could not meet with grade B of discharge standard of pollutants for municipal wastewater treatment plant in China (GB/T18918-2002) at Gas/water ratio of 10/1, 14/1 and 24/1, while could meet with grade B of this standard at Gas/water ratio of 5/1 and could meet with grade A of this standard at Gas/water ratio of 7/1.
- The Gas/water ratio in range 5/1 to 24/1 did not significantly affect COD and $\text{NH}_4^+\text{-N}$ removal efficiencies.
- In overall, the results showed that the Gas/water ratio of 7/1 is optimal for simultaneous organics and nutrients removal.

REFERENCES

- Al-Ghusain, I.A., O.J. Hao, J.M. Chen, C.F. Lin, M. Kim and A. Torrents, 1994. Biological fixed-film systems: A literature review. *Wat. Env. Res.*, 64 (4): 336-354.
- Anderson, I.C. and J.S. Levine, 1986. Relative rates of nitric oxide and nitrous oxide production by nitrifiers, denitrifiers and nitraterespirers. *Appl. Env. Microbiol.*, 51: 938-945.
- Andreottola, G., P. Foladori and M. Ragazzi, 2000b. Upgrading of a small wastewater treatment plant in a cold climate region using a Moving Bed Biofilm Reactor (MBBR) system. *Water Sci. Technol.*, 41: 177-185.
- Andreottola, G., P. Foladori, M. Ragazzi and F. Tatano, 2000a. Experimental comparison between MBBR and activated sludge system for the treatment of municipal waste water. *Water Sci. Technol.*, 41: 375-382.
- Andreottola, G., P. Foladori, M. Ragazzi and R. Villa, 2002. Dairy wastewater treatment in a moving bed biofilm reactor. *Water Sci. Technol.*, 45(12): 321-328.
- Anthonisen, A.C., R.C. Loehr, T.B.S. Prakasam and E.G. Srinath, 1976. Inhibition of Nitrification by ammonia nitrous acid. *J. Water Pollut. Con. F.*, 48(5): 825-835.
- Bae, W., S. Baek, J. Chung and Y. Lee, 2001. Optimal operational factors for nitrite accumulation in batch reactors. *Biodegradation*, 12: 359-366.

- Bernet, N., P. Dangcong, J.P. Delgenes and R. Moletta, 2001. Nitrification at low oxygen concentration in biofilm reactor. *J. Environ. Eng. ASCE.*, 127: 266-271.
- Botrous, A.E.F., M.F. Dahab and P. Mihaltz, 2004. Nitrification of high strength ammonium wastewater by a fluidized-bed reactor. *Water Sci. Technol.*, 49: 65-71.
- Canziani, R., V. Emondi, M. Garavaglia, F. Malpei, E. Pasinetti and G. Buttiglieri, 2006. Effect of oxygen concentration on biological nitrification and microbial kinetics in a cross-flow membrane bioreactor (MBR) and moving-bed biofilm reactor (MBBR) treating old landfill leachate. *J. Membrane Sci.*, 286: 202-212.
- Chen, J.M., O.J. Hao, I.A. Al-Ghusain and C.F. Lin, 1995. Biological fixed-film systems: A literature review. *Wat. Env. Res.*, 67(4): 450-469.
- Ciudad, G., O. Rubilar, P. Munoz, G. Ruiz, R. Chamy, C. Vergara and D. Jeison, 2005. Partial nitrification of high ammonia concentrates on waste water as a part of a shortcut biological nitrogen removal process. *Process Biochem.*, 40: 1715-1719.
- Delenfort, E. and P. Thulin, 1997. The use of Kaldnes suspended carrier process in treatment of wastewaters from the forest industry. *Water Sci. Tech.*, 35(2-3): 123-130.
- Falletti, L. and L. Conte, 2007. Upgrading of activated sludge wastewater treatment plants with hybrid moving-bed biofilm reactors. *Ind. Eng. Chem. Res.*, 46: 6656-6660.
- Hagedorn-Olsen, C., I.H. Moller, H. Tottrup and P. Harremoës, 1994. Oxygen reduces denitrification in biofilm reactors. *Water Sci. Technol.*, 29: 83-91.
- Halling, S.B. and S.G. Jorgensen, 1993. Removal of Nitrogen Compounds from Waste Water. Elsevier Publishers, Amsterdam.
- Helmer, C., S. Kunst, S. Juretschko, M.C. Schmid and M. Wagner, 1999. Nitrogen loss in a nitrifying biofilm system. *Water Sci. Tech.*, 39(7): 13-21.
- Helness, H., 2007. Biological phosphorous removal in a movingbed biofilm reactor. Ph.D. Thesis, Norwegian University of Science and Technology, Norway.
- Huang, J., M. Chen and P. Davis, 1992. Biological fixed-film systems: A literature review. *Water Environ. Res.*, 64(4): 359-378.
- Jahren, S.J., J.A. Rintala and H. Odegaard, 2002. Aerobic moving bed biofilm reactor treating thermomechanical pulping whitewater under thermophilic conditions. *Water Res.*, 36: 1067-1075.
- Kelly, G., 1996. Environmental Engineering. McGraw Hill Publishing House: Maidenhead, England.
- Lie, E. and T. Welander, 1994. Influence of dissolved oxygen and oxidation-reduction potential on the nitrification rate of activated sludge. *Water Sci. Technol.*, 30: 91-100.
- Lindemann, J. and U. Wiesmann, 2000. Single-disc investigations on nitrogen removal of higher loads in sequencing batch and continuously operated RDR systems. *Water Sci. Technol.*, 41: 77- 84.
- Maurer, M., C. Fux, M. Graff and H. Siegrist, 2000. Moving Bed Biological Treatment (MBBT) of municipal wastewater: Denitrification. *Water Sci. Technol.*, 43: 337-344.
- Odegaard, H., B. Rusten and T. Swestrum, 1994. A new moving bed biofilm reactor applications and results. *Water Sci. Tech.*, 29(10-11): 157-165.
- Odegaard, H., B. Gisvold and J. Strickland, 2000. The influence of carrier size and shape in the moving bed biofilm process. *Water Sci. Technol.*, 41(4-5): 383-391.
- Okabe, S. and Y. Watanabe, 2000. Structure and function of nitrifying biofilms as determined by in situ hybridization and the use of microelectrodes. *Water Sci. Technol.*, 42: 21-32.
- Park, H.D. and D.R. Noguera, 2004. Evaluating the effect of dissolved oxygen on ammonia-oxidizing bacterial communities in activated sludge. *Water Res.*, 38: 3275-3286.
- Pastorlli, G., R. Canzizni and A. Rozzi, 1999. Phosphorus and nitrogen removal in MBBRs. *Water Sci. Tech.*, 40(4-5): 169-176.
- Ritchie, G.A.F. and D.J.D. Nicholas, 1972. Identification of the sources of nitrous oxide produced by oxidative and reductive processes in *Nitrosomonas* European. *Biochem. J.*, 126: 1181-1191.
- Rittmann, B.E. and P.L. McCarty, 2001. Environmental Biotechnology: Principles and Applications. McGraw-Hill, New York, pp: 470-4.
- Ruiz, G., D. Jeison and R. Chamy, 2003. Nitrification with high nitrite accumulation for the treatment of wastewater with high ammonia concentration. *Water Res.*, 37: 1371-1377.
- Rusten, B., A. Broch-Due and T. Westrum, 1994. Treatment of pulp and paper industry wastewater in novel MBBR. *Water Sci. Tech.*, 30(3): 161-171.
- Rusten, B., L. Hem and H. Odegaard, 1995. Nitrification of municipal wastewater in novel moving bed biofilm reactors. *Water Environ. Res.*, 67: 75-86.
- Rusten, B., J.G. Siljudalen and H. Strand, 1996. Upgrading of abiological- chemical treatment plant for cheese factory waste water. *Water Sci. Tech.*, 34 (11): 41-49.
- Rusten, B., O. Sehested and B. Svendsen, 2000. Pilot testing and preliminary design of MBBRs for nitrogen removal at the FREVAR wastewater treatment plant. *Water Sci. Tech.*, 41(4-5): 13-20.
- State Environmental Protection Administration, 2002. Water and Wastewater Monitoring and Analysis Methods. 4th Edn., China Environmental Science Press, China.

- Wang, J. and N. Yang, 2004. Partial nitrification under limited dissolved oxygen conditions. *Process Biochem.*, 39: 1223-1229.
- Wiesmann, U., 1994. Biological Nitrogen Removal from Wastewater. In: A. Fiechter (Ed.), *Advances in Biochemical Engineering/Biotechnology*. Springer-Verlag, Berlin-Heidelberg.
- Xiao, L.W., M. Rodgers and J. Mulqueen, 2007. Organic carbon and nitrogen removal from a strong wastewater using a denitrifying suspended growth reactor and a horizontal-flow biofilm reactor. *Bioresour. Technol.*, 98: 739-744.
- Zumft, W.G., 1997. Cell biology and molecular basis of denitrification. *Microbiol. Mol. Biol. Rev.*, 61: 533-616.