EVALUATION OF HOLLOW FIBER MEMBRANE BIOREACTOR EFFICIENCY FOR MUNICIPAL WASTEWATER TREATMENT

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ABSTRACT
The membrane bioreactor technology has been proven to be a single step process in efficient treatment of wastewater, either directly or after pretreatment by reverse osmosis. In this study, a pilot scale experiment was studied to treat a synthetic municipal wastewater sample. The aerobic reactor with a submerged membrane used in this work was continuously aerated for organic matter oxidation, nitrification and phosphorous uptake as well as for fouling control. The mixed liquor was recycled from the aerated zone to the anoxic zone for denitrification. The membrane had a nominal pore size of 0.1 µm and a filtration area of 4.0 m^2. The performance of submerged membrane bioreactor was examined in order to determine the removal efficiency of organic compounds and nitrogen in different solid retention times (10, 20, 30, and 40 days) under a continuous inflow of the synthetic municipal wastewater. Results indicated that the submerged membrane bioreactor could efficiently remove the pollutants. Average removal rates of chemical oxygen demand, total Kejeldahl nitrogen removal, total nitrogen and phosphorous reached to as high as 99.3%, 98.1%, 85.5%, and 52%, respectively. Furthermore, concentrations of nitrate and nitrite in the last stage were well reduced and reached to 5.3 and 0.047 mg/L, respectively.

Key words: Membrane bioreactor, wastewater treatment, solid retention time

INTRODUCTION
Due to diminishing water supplies and increasing population, wastewater reclamation is becoming necessary throughout the world to conserve natural water resources used for drinking water supply. The membrane bioreactor (MBR) is a leading edge technology currently being used in countries around the world for water reclamation. Due to advances in technology and declining costs, the application of MBR technology for water reclamation has sharply increased over the past several years (Adham *et al*., 2004).

Membrane bioreactor is a biological wastewater treatment process that uses membrane to replace the gravitational settling of the conventional activated sludge process for the solid–liquid separation of sludge suspension. MBRs, in which biomass is strictly separated by a membrane, offer several advantages over the conventional activated sludge process, including a higher biomass concentration, reduced footprint, low sludge production, and better permeate quality (Yamamoto *et al*., 1989; Van Dijk *et al*., 1997).

The technology of MBRs and their application in domestic wastewater treatment as well as industrial wastewater has recently been paid closer attention due to demands to deliver effluents to higher standards and more reliable quality. They can be broadly defined as systems integrating biological degradation of wastewater with Membrane filtration. They have proven to be effective in removing organic and inorganic contaminants as well as biological entities (Laera *et al*., 2007). As a result, MBR technology is accepted as a reliable and advanced option for wastewater treatment, which can replace the conventional biological methods (Seong-Hoon Yoon *et al*., 2006).

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MBRs are categorized into a cross flow type and a submerged type according to allocation of the membrane module. In a cross flow MBR, a membrane module is allocated outside a bioreactor and mixed liquor is driven into the membrane module by recirculation pumps. The cross flow membrane generally has a high flux and an easy-maintenance. In a submerged MBR, a membrane module is submerged into a bioreactor and mixed liquor is generally suctioned from the effluent side, reducing the operating cost since the cake layer is removed by the uplifting flow of bubbling air and mixed liquor and leading to decrease of pumping energy costs. The mean cell residence time (SRT) can influence biomass characteristics in activated sludge systems, and the concentration of mixed liquor suspended solids (MLSS) in the bioreactor tends to increase at higher SRT.

In MBR, very efficient solid/liquid separation is provided, thus higher MLSS can be maintained compared with conventional activated sludge systems. However, treatment efficiency is not linearly dependent on biomass concentration because the specific biological activity can be reduced at substrate-deficient states (Soong-Soo *et al*., 2005). In a few studies in MBR systems, it has been found that biomass production can be limited by proper operational decisions (Lu *et al*., 2001; Xing *et al*., 2003). It has been reported that total nitrogen cab be removed in an MBR by integrating an anoxic bioreactor (Rosenberger *et al*., 2002; Yoon *et al*., 2004), and always pre-denitrification is a preferred framework which is as a result of the endogenous use of organic matters in the wastewater. This will provide a substantial saving in the chemical (supplementary organics) cost. By recycling high flow of returned MLSS more nitrate would be returned back to the anoxic region of the tank and denitrification will take place and escaping of nitrate will be prevented. Fan *et al*., 2000 reported that perfect nitrification could be achieved in MBR systems.

The aeration condition in the MBR systems is different from that of the conventional activated sludge (CAS) (Teck Wee Tan *et al*., 2007). Usually, high aeration rate is always used in the MBR to provide high mixing and also to control membrane fouling (Liu *et al*., 2000; Germain *et al*., 2005). Therefore, in the MBR systems, the DO concentration can easily fluctuate above 4 mg O_2/L (Chu and Li, 2005; Yoon *et al*., 2004). Application of pre-coagulation/sedimentation processes will prevent membrane fouling in subsequent MBRs and also removal of phosphorus can be enhanced (Watanabe *et al*., 2006). When the MBR system is operating at sludge age between 40 and 80 days, physical properties of the sludge such as dewaterability, filterability, and settleability would be improved (Pollice *et al*., 2007).

In recent years, MBR aims application has to be operated in long sludge retention time (SRT) condition to minimize the excess sludge production (Gander *et al*., 2000; Rosenberger *et al*., 2002; Innocenti *et al*., 2002; Sun *et al*., 2005).

The present work reports results obtained from investigating the effects of different SRTs on removal rates of chemical oxygen demand (COD), total nitrogen (TN) and total phosphorous (TP) from a synthetic municipal wastewater by use of a pilot scale submerged membrane bioreactor (SMBR) system.

MATERIALS AND METHODS

MBR system, raw wastewater and operational conditions

The experimental set-up consisted of a modified Ludzack-Ettinger (MLE) type SMBR process. Fig. 1 shows the schematic diagram of the submerged MBR system used in this study. The system composed of an activated sludge bioreactor having a submerged microfiltration membrane module. The bioreactor was separated into two sections. One was an anoxic rector for denitrification (A-tank) and the other an oxic reactor (B-tank) for organic matter removal and nitrification. The effective volumes of these two sections were 36 L and 48 L, respectively. For providing the best condition of denitrification and homogenization of mixed liquor, a top-mount stirrer was used in the anoxic zone. Oxygen demand was supplied by four air pumps attached to the diffusers inserted at the bottom of the membrane. DO concentration in the B-tank was kept above 4.5 mg/L while in the A-tank it was generally below 0.5 mg/L. The operating temperature and pH were adjusted at 22±2°C and 6.5-8.0. Transmembrane pressure was measured and controlled by using an analytical manometer. The amount of vacuum applied for suction was 0.15 bars. The microfiltration hollow fiber membrane module used for this study was manufactured from Zenon Company of Canada. This membrane was submerged in the oxic zone of MBR for wastewater treatment. Technical data is presented in Table 1.

Table 1: The specifications of the hollow fiber membrane

Physical property	Specifications
Raw material	Polypropylene
Inside diameter (µm)	320
Pore size (µm)	0.1
Number of layers	3
External shape	Hollow fiber
Outside diameter (µm)	400
Pore density (%)	40-50
Length × width (mm)	450 × 250

* Data taken from Zenon Co.

The membrane module was placed in the center of the B-reactor to ensure maximum contact with the coarse air bubbles and for alleviating the fouling phenomenon commonly encountered in MBR. Seeding of the wastewater in MBR system was fulfilled by using the sludge from the Tehran West Residual District wastewater treatment plant. In this study, MBR system was fed with a synthetic wastewater for about 90 days. This synthetic wastewater contained appropriate amounts of glucose ($C_6H_{12}O_6$) and useful salts like KH_2PO_4, $(NH_4)_2SO_4$, $MgSO_4$, $CaCl_2$, $FeCl_3$, $MgSO_4$ and $NaHCO_3$. Characteristics of the synthetic wastewater were as follows: pH=7.3, COD = 500 mg/L, NH_4-N=42 mg/L, phosphorous=12 mg/L and total alkalinity=80 mg/L as $CaCO_3$. Sampling for determination of system efficiency was started when MLSS concentration reached to about 7500 mg/L. This period was prolonged about 30 days, and in this time no sludge was disposed.

The list of operational conditions is presented in Table 2. The MBR system was operated with a flow rate of 96 L/d.The hydraulic retention times (HRTs) of anoxic tank and oxic tank were 2.6 and 12 h,respectively .For removal of nitrate and performing denitrification process the mixed liquor suspended solid (MLSS) was returned to the anoxic reactor at a rate of 250%. Flux of the membrane and organic loading rate were 1 L/m^2.h and 0.5 Kg COD/m^3.d, respectively. To minimize membrane fouling, filtration was performed in an intermittent fashion of alternating 10 min suction and 4 min pause.

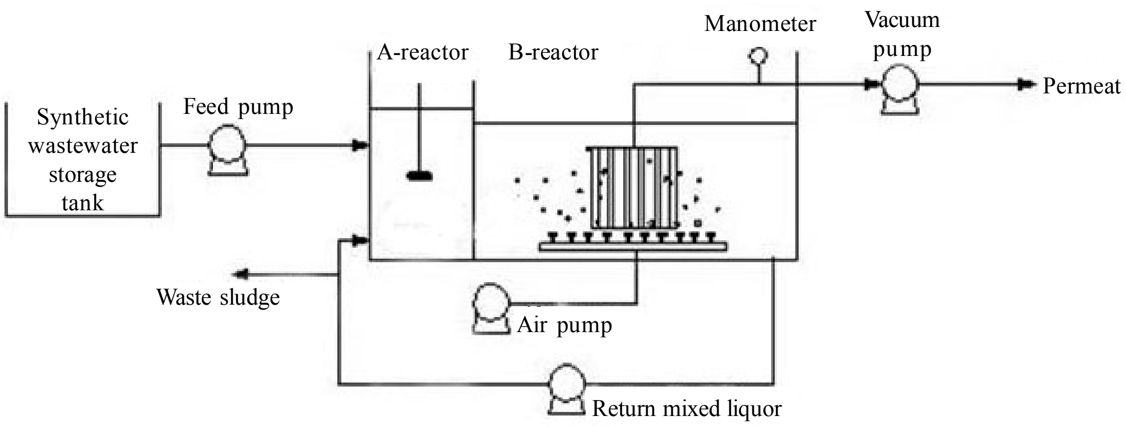

Fig. 1: Schematic of SMBR system (Modified Ludzack-Ettinger)

Table 2: Operating conditions of the submerged MBR with different SRTs (days)

Parameters	SRT (day)			
	10	20	30	40
Influent flow (L/d)	96	96	96	96
MLSS (g/L)	8	9	11.5	13.5
MLVSS (g/L)	6	7	9.5	11.5
Waste sludge (L/d)	4.8	2.4	1.2	0.6

*HRT – hydraulic retention time
*The amount of MLSS and MLVSS are based on arithmetic mean in each run

Analytical methods

Samples of influent synthetic municipal wastewater and permeate were analyzed every day and examined for soluble COD (5220 B), TKN (4500-N_{org}-B), N-NO_2 (4500-NO_2-B), N-NO_3 (4500-NO_3-C) and TP (4500-P-C). All analyses were performed according to the Standard Methods (APHA, 1995). Concentration of DO, pH and temperature were measured by Dissolved Oxygen Meter (YSI model 50B), pH meter and a thermometer, respectively.

The system was tested for eight days in each run after reaching to the steady state. Before each run, COD of effluent stream was measured daily and the trend of its change versus time was drawn. Steady state could be considered as the time when the curve of COD reached to a stationary condition.

RESULTS

Fig. 2 shows the curve of biomass growth versus time for all operating conditions. Applying different SRTs had resulted in different COD removal rates as can be seen in Fig. 3. The data needed for depicting this figure are presented in Table 3. Fig. 4 shows the TKN removal rates obtained at different SRTs and the related data are presented in Table 4.

The concentrations of nitrite and nitrate in feed and permeate at different SRTs could be seen in Figs. 5 and 6, respectively. Table 5 shows the percent of total nitrogen removal obtained at different SRTs. Finally, Fig. 7 shows the TP removal rates obtained at different SRTs. The data used for depicting this figure are presented in Table 6.

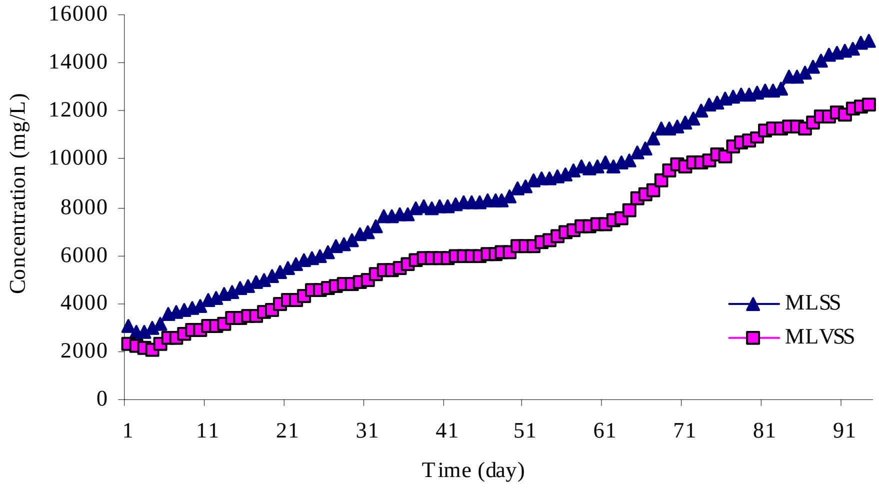

Fig. 2: Biomass growth with time for all operating conditions

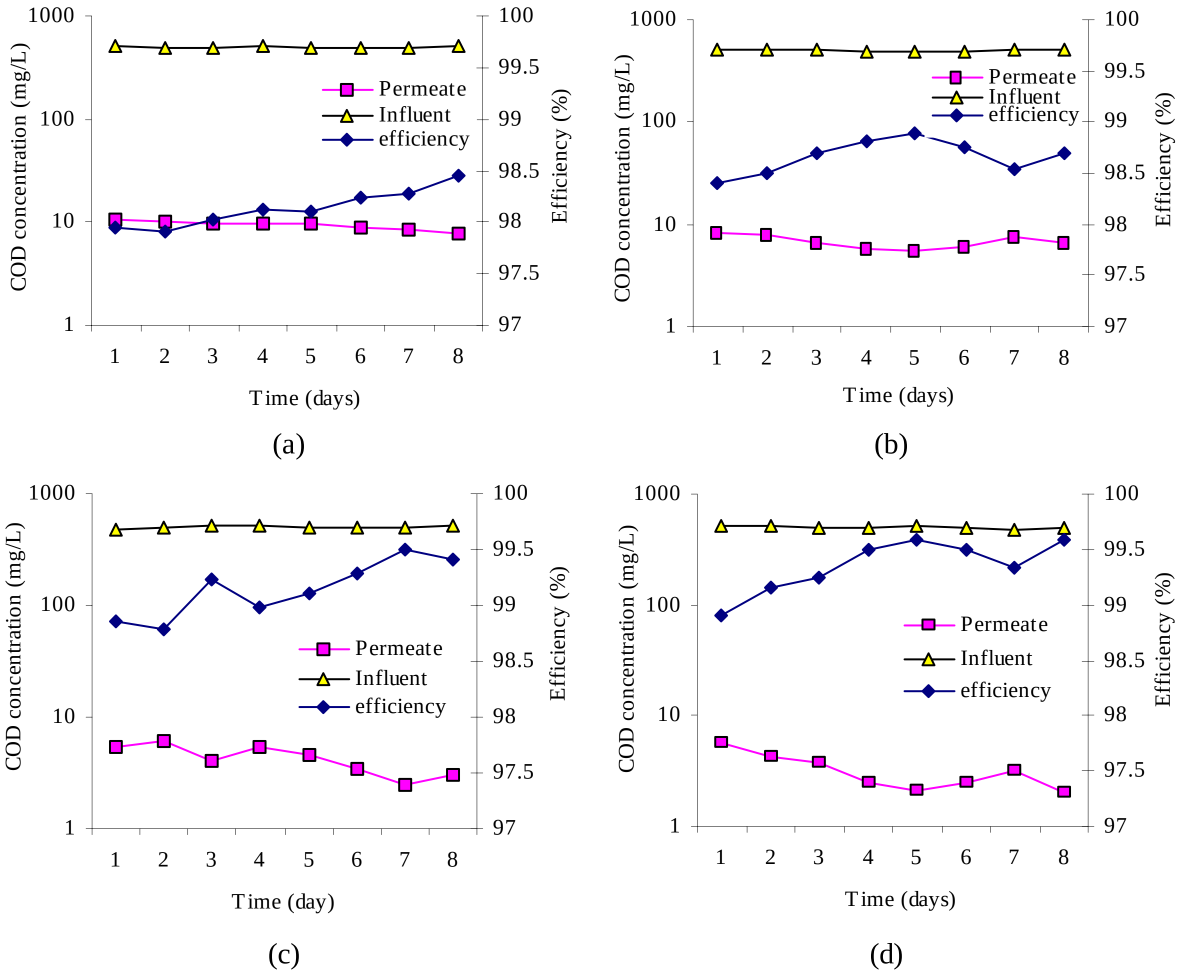

Fig. 3: Soluble COD removal rates at different SRTs (days); a (10), b (20), c (30), d (40)

Table 3: Percent of soluble COD (SCOD) removal at different SRTs (days)

SCOD (average)	SRT (day)			
	10	20	30	40
Influent (mg/L)	500	502.7	503.8	500.5
Permeate (mg/L)	9.33	6.76	4.3	3.5
Removal (%)	98.13	98.65	99.15	99.35

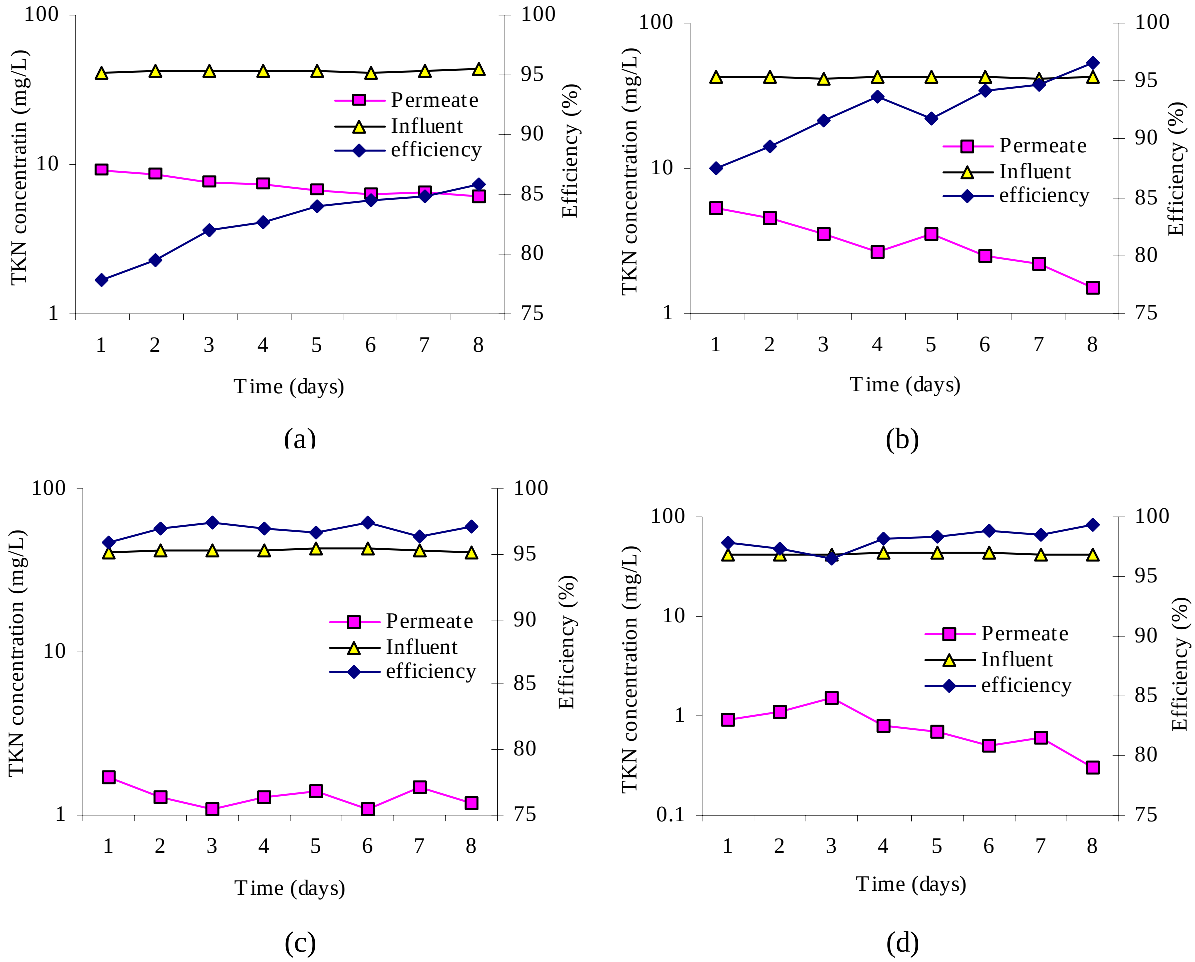

Fig. 4: TKN removal rates at different SRTs (days); a (10), b (20), c (30), d (40)

Table 4: Percent of Total Kejeldahl Nitrogen (TKN) removal in different SRTs (days)

TKN (average)	SRT (day)			
	10	20	30	40
Influent (mg/L)	42.2	42.4	41.87	42.32
Permeate (mg/L)	7.3	3.23	1.32	0.8
Removal (%)	82.67	92.40	96.84	98.12

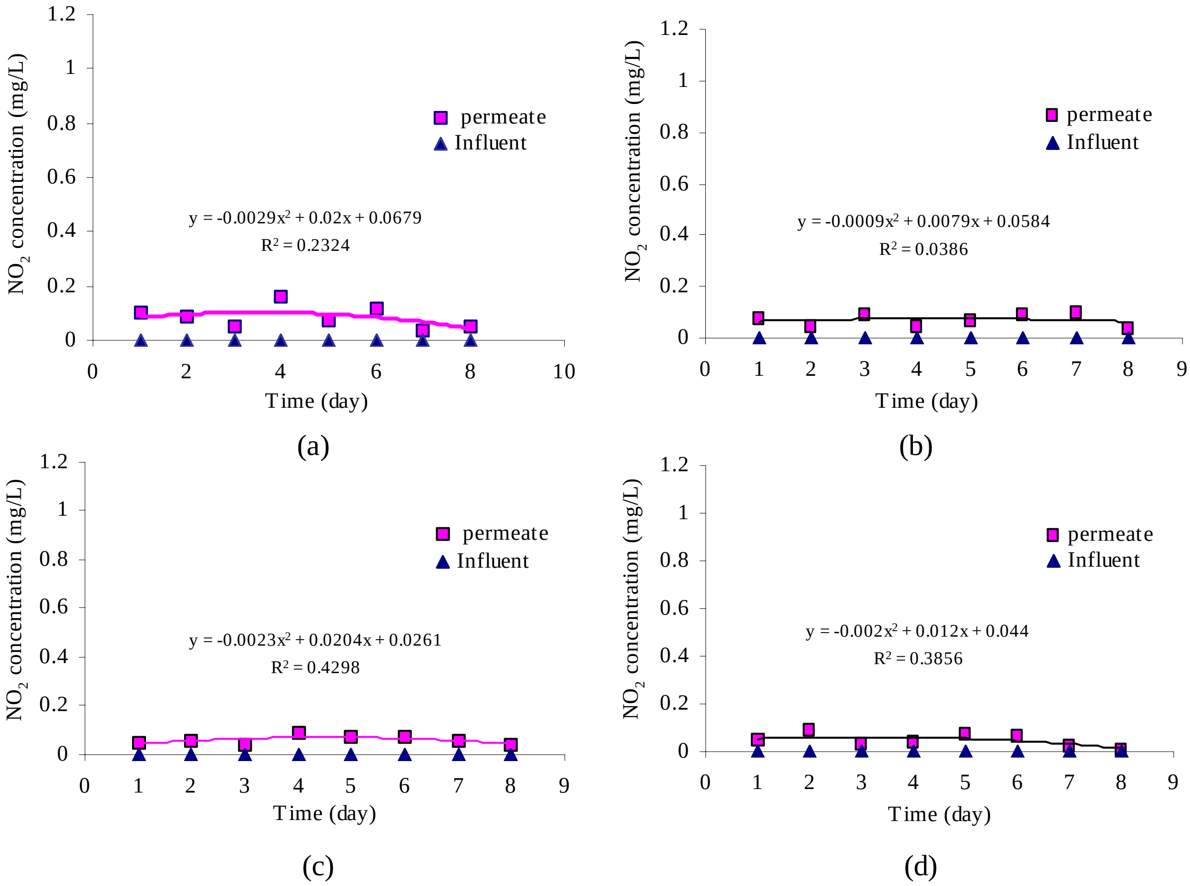

Fig. 5: Concentrations of nitrite in feed and permeate at different SRTs (days); a (10), b (20), c (30), d (40)

Table 5: Percent of total nitrogen (TN) removal at different SRTs (days)

TN (average)	SRT (day)			
	10	20	30	40
Influent (mg/L)	42.2	42.4	41.87	42.32
Permeate (mg/L)	15.32	9.32	7.23	6.16
Removal (%)	63.70	78.01	82.73	85.44

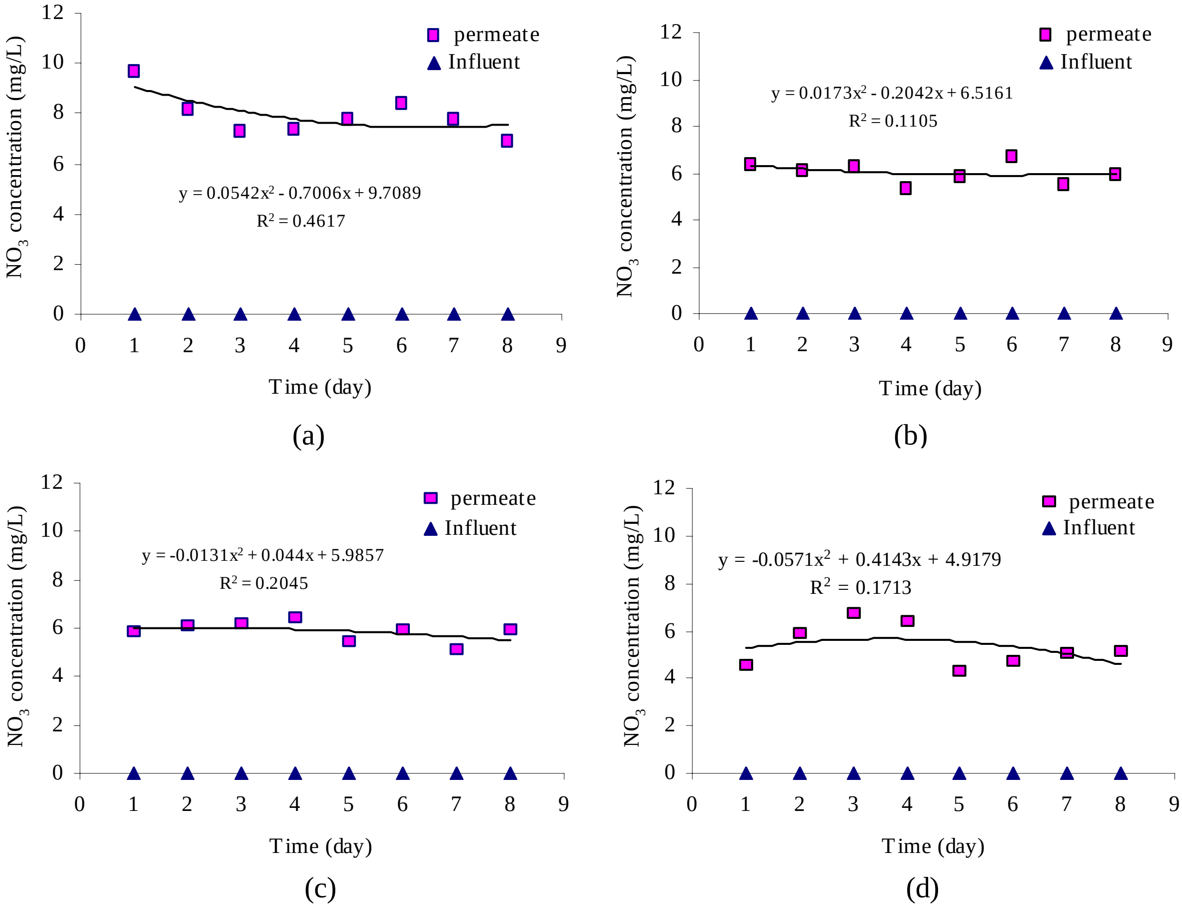

Fig. 6: Concentrations of nitrate in feed and permeate at different SRTs (days); a (10), b (20), c (30), d (40)

Table 6: Percent of phosphorous removal (TP) removal at different SRTs (days)

TP (average)	SRT (day)			
	10	20	30	40
Influent (mg/L)	12.35	12.52	12.65	12.76
Permeate (mg/L)	8.92	8.38	7.22	6.11
Removal (%)	27.73	32.97	42.89	52.10

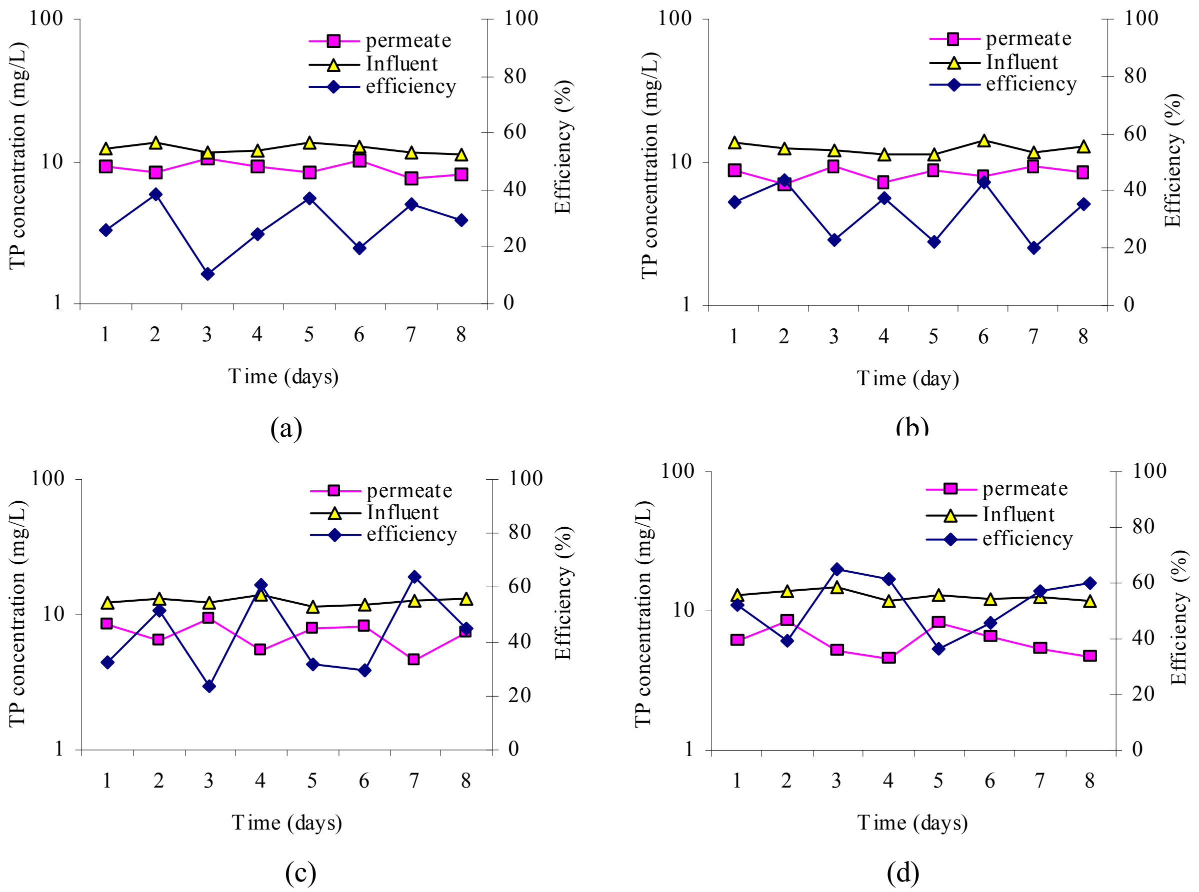

Fig. 7: Phosphorous removal rates at different SRTs (days); a (10), b (20), c (30), d (40)

DISCUSSION

As shown in Fig. 2, MLSS and MLVSS concentrations increased with SRT increase. In the first 1 to 5 days, the concentrations of MLSS and MLVSS decreased and this result was in agreement with Coello Oviedo *et al*., 2003 who demonstrated that the largest decrease in MLSS occurs in the first 4 days possibly as a result of the death of higher microorganisms due to the lack of organic material in the reactor. The MLVSS over MLSS concentration ratio (MLVSS/MLSS ratio) in an MBR denotes the organic component in the sludge and Variations in this ratio indicate a change in the biomass component. It was indicated that in all operating conditions of this study this ratio was remained almost constant (in the range of 0.75–0.85). This result was in accordance with Marija Vukovic *et al.,* 2006 who analyzed the activated sludge process in an MBR under starvation conditions and demonstrated that during experimental runs, the ratio of MLVSS/MLSS was almost constant (in the range of 0.76–0.85) over the whole experimental period. This result also was in agree with Rosenberger *et al.,* 2002 who demonstrated that no decrease in the volatile fraction was observed with the MLVSS/MLSS ratio remaining constant at approximately 0.75 throughout the continuous three years operation with 18-20 g/L MLSS and 1.7 kg COD/m^3.day of domestic wastewater. The biomass concentration at the prolonged SRT in MBR was higher than the conventional activated sludge process with settling limitation normally restricted to 6 g/L concentration (Defrance and Jaffrin, 1999; Smith *et al*., 2003).

The results from Fig. 3 and Table 3 shows that SMBR system can remove SCOD efficiently, This result was in agreement with Ueda *et al.*, 1999; Cote *et al.*, 1997, 1998, Ahn *et al.*, 2003, Patel *et al*. 2005 and Rosenberger *et al.*, 2005 who investigated the influence of different SRTs on different operating parameters of MBR and all reported a COD removal rate in this range. The mean concentration of permeate SCOD decreased as SRT increased and in SRT of 40 days the SCOD in permeate was lower than 5 mg/L. This result was in agreement with S. L. Khor *et al*., (2006) that examined a submerged membrane bioreactor in different SRT conditions and reported that in 5 days, 10 days and prolonged SRT MBRs, the overall organic degradation efficiencies in this system were 97.46%, 98.02% and 98.94% respectively. This high removal efficiency implies that in the membrane bioreactor system, organic matter can be degraded in high level, because of high concentration of biomass. This overall organic degradation efficiency was attributed by the biological degradation and membrane filtration. The membrane filtration played a significant role in maintaining high and stable organic removal efficiency. These results for COD removal were better than removal efficiencies that obtained from other systems such as activated sludge and SBR. For example, COD removal efficiency in SBR process was reported by Mahvi *et al*., (2004, 2008) to be 94% in the best condition.

Because the synthetic feed wastewater was prepared daily, concentrations of NO_3 and NO_2 in feed wastewater were remained zero in all operating conditions. According to these results, the mean concentrations of NO_3 and NO_2 in MBR permeate were decreased as SRT increased. These results were in accordance with the results reported by Samer Adham *et al*., (2004).

According to Fig. 4 and Table 4, the mean TKN removal efficiency in all operating conditions was in the range of 82-98%. These results were in agreement with P.Battistoni *et al*., (2006) who reported the removal efficiencies of TKN equal to 97 and 96%, respectively in 21 and 14 days SRTs. As shown in Fig. 4, as SRT increased the TKN concentration of permeates decreased.

Because of synthetic inorganic material used instead of natural nutrients exist in wastewaters; all of the TKN amounts that had been measured in the influent wastewater were of NH4-N type. Besides, as total concentrations of NO_3 and NO_2 in the influent wastewater were equal to about zero, TN concentration in the feed wastewater was equal to TKN concentration. However this was not true for permeate, because TN concentration in permeate was equal to the sum of TKN, NO_3 and NO_2 concentration, due to bacterial activity.

As shown in Table 5, the efficiency of TN removal in the best condition (SRT=40 day) was 85.4%. This result was in accordance with Fu Guokai *et al*., (2007) who investigated removal of organic matter and nitrogen in municipal wastewater by a new submerged membrane bioreactor and reported that average rate of TN removal in a new MBR was equal to 80% in 40 days SRT. This showed that the removal of nitrogen could be attributed mainly to the action of simultaneous nitrification and denitrification happening in the MBR. Results showed that the nitrogen compounds in the effluent appeared mostly in the form of nitrate nitrogen as SRT increased, and this indicated that nitrification was perfectly complete. Therefore, TN removal would depend mainly to the degree of the denitrification in the anoxic reactor.

As it was expected, the removal efficiency of phosphorous in all operating conditions was not very high, because the system did not consist of an anaerobic reactor. But by adding conventional coagulant salts to anoxic or oxic reactor, the removal efficiency of phosphorous may be increased.

For conclusion, this study surveyed the effect of different solid retention times in removal of COD, TN and TP. The results clearly showed that the SMBR that used anoxic and oxic reactors was able to achieve very good organic removal efficiencies whatever the SRT was. Furthermore, the results showed that this system can remove TKN and TN very efficiently and as expected, concentrations of NO_3 and NO_2 in permeate were always low. With regard to the performance of this system in removing TP, it could be claimed that the system had also removed this nutrient relatively good although this system was not equipped with the required anaerobic zone. Nevertheless, TP removal efficiency of more than 50% was only achievable when SRT was 40 days. Thus, addition of metal salts would be necessary for meeting strict standards of TP discharge. Finally, it could be claimed that membrane bioreactor system that uses anoxic/oxic reactors may have a good role in removal of organic matter and nutrient compounds and compared with other activated sludge processes, quality of effluent is better and thus it can be used successfully for wastewater treatment and water recycling.

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