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Impact of nitrogen loading rates on treatment performance of domestic wastewater and fouling propensity in submerged membrane bioreactor (MBR)

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HIGHLIGHTS

- Higher nitrogen loading rate adversely affected nutrient removal efficiency.
- Soluble EPS concentration increased with increasing ammonium nitrogen concentration.
- High soluble EPS concentration caused higher cake layer membrane resistance.
- Membrane filtration performance deteriorated with increasing nitrogen loading rate.

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ABSTRACT

In this study, performance of laboratory-scale membrane bioreactor (MBR) was evaluated in treating high strength domestic wastewater under two nitrogen loading rates (NLR) i.e., 0.15 and 0.30 kg/m^3/d in condition 1 and 2, respectively, while organic loading rate (OLR) was constant at 3 kg/m^3/d in both conditions. Removal efficiencies of COD were above 95.0% under both NLR conditions. Average removal efficiencies of ammonium nitrogen (NH_4^+–N), total nitrogen (TN) and total phosphorus (TP) were found to be higher in condition 1 (90.5%, 74.0%, and 38.0%, respectively) as compared to that in Condition 2 (89.3%, 35.0%, and 14.0%, respectively). With increasing NLR, particle size distribution shifted from narrow (67–133 μm) towards broader distribution (3–300 μm) inferring lower cake layer porosity over membrane fibers. Soluble extracellular polymer substance (sEPS) concentration increased at higher NLR due to biopolymers released from broken flocs. Higher cake layer resistance (R_c) contributed towards shorter filtration runs during condition 2.

1. Introduction

New and stringent wastewater effluent discharge standards have necessitated the use of membrane bioreactors (MBRs), which is the most promising and reliable technology for wastewater treatment and reuse (Huang et al., 2008; Wang et al., 2008). The expected annual growth rate for global market of MBR is 10.5%, and it is increasing in value from 296.0 million dollars in 2008 to 488.0 million dollars by 2013 but growth rates of MBR systems are not the same for all world regions (Kraume and Drews, 2010). Because of the high quality treated effluents produced by MBRs, they are being considered a future replacement for conventional activated sludge systems especially in those regions of the world where water scarcity problems force to increase the reuse of high quality treated wastewater (Bolzonella et al., 2010).

Although the capital and energy costs of MBR has been reduced but fouling which is major problem for MBRs exists as a black box due to the complex nature of fouling layers (Meng et al., 2010). In spite of many efforts to control membrane fouling in MBRs, consistent and detailed solutions in real cases as well as which are economically feasible are still not clear (Wu and Fane, 2012). Membrane fouling is attributed to cake layer, gel layer and pore blocking. Many researchers revealed the cake layer as main contributor for membrane fouling in MBR (Huang et al., 2008; Khan et al., 2009; Meng et al., 2008). Operating conditions, biomass characteristics and membrane properties are the main factors that affect the performance of MBR (Wang et al., 2009). Mixed liquor in MBR is a mixture of biomass, feed water components and metabolic products produced during different biologi-

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cal reactions (Jiang et al., 2003). Fouling is affected by mixed liquor properties such as mixed liquor suspended solids (MLSS) concentration, viscosity, floc size, and extracellular polymeric substance (EPS).

EPS which is considered as major foulant exist in two forms, bound and soluble/colloidal. Bound EPS comprise of substances attached around the cells (Andreadakis, 1993). Soluble EPS also known as soluble microbial products (SMP) are bacterially produced polymers, lysis products and hydrolysis products or products of dissolution of bound EPS. Main components of EPS and SMP are proteins and polysaccharides (Wu and Huang, 2009).

Performance of MBR is also affected by loading rates. Åhl et al. (2006) observed higher fouling rates at high organic loading rates (OLR) of 7.8 kg/m^3/d as compared to low loading rates 2.3 kg/m^3/d and found good COD removal efficiency at low OLR. In order to control the fouling, the effect of loading rates on sludge characteristics must be investigated. In this context the objective of this study was to investigate the effect of nitrogen loading rates on membrane filtration and treatment performance in submerged MBR. The influence of mixed liquor properties on membrane fouling propensity was also examined.

2. Methods

2.1. Reactor setup and operating parameters

A laboratory-scale submerged membrane bioreactor with working volume of 14 L was used in this study. The reactor was made of acrylic and it was divided into three compartments with two porous baffle sheets. In the central compartment of the reactor hollow fiber (HF) membrane module (Mitsubishi Rayon, Japan) with filtration area of 0.2 m^2 and nominal pore size of 0.1 μm was submerged as shown in Fig. 1. A suction pump (Master Flex, Cole–Parmer, USA) was used to extract the membrane filtrate in intermittent mode with 10 min filtration and 2 min relaxation cycle. The MBR was operated under two conditions i.e., condition 1 and condition 2. Duration of condition 1 was 95 days and condition 2 was 65 days. All operating parameters were kept constant in both conditions except NLR which was doubled in condition 2. To supply oxygen to microorganisms and for effective membrane scouring diffused aeration system was used. The solids retention time (SRT) was maintained at 30 days while hydraulic retention time (HRT) was kept at 8 h. The aeration rates were maintained at 7 L/min (0.42 m^3/h) (4 L/min in membrane compartment and 3 L/min in side compartments). DO was maintained in the range of 3–5 mg/L.

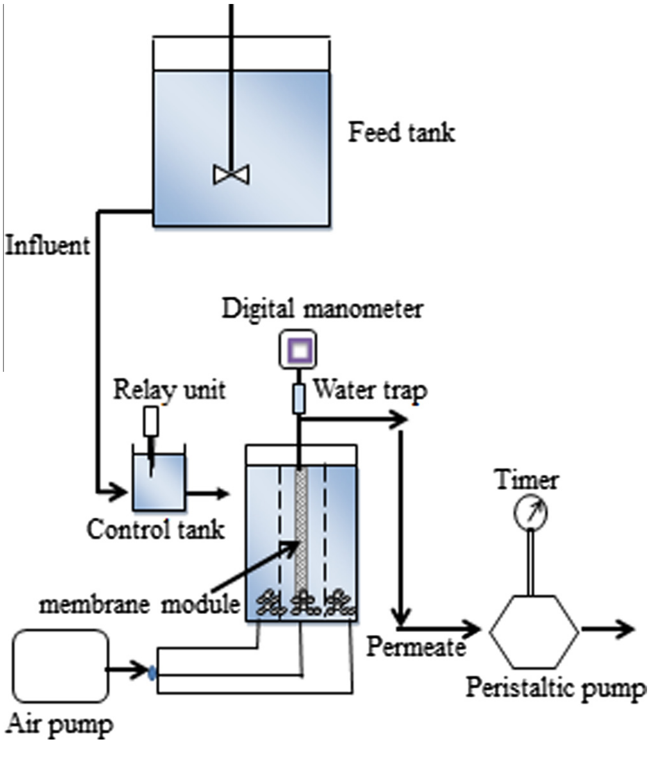

Fig. 1. Schematic of the MBR system.

2.2. Wastewater composition and seed sludge

For this study, seed sludge was taken from aeration tank of Sewage Treatment Plant (STP), Sector I-9, Islamabad, Pakistan, with initial MLSS concentration of 2000 mg/L. The sludge was acclimatized in sequencing batch reactor (SBR) with synthetic wastewater for 60 days to attain the required MLSS concentration of 8000–9000 mg/L and to maintain COD removal efficiency above 80% before shifting to the MBR tank. High strength synthetic wastewater was used for this study. Organic-loading rate (OLR) was 3 kg/m^3/d (COD 1000 mg/L) and it was kept constant in both conditions. NLR was maintained at 0.15 kg/m^3/d (COD:N = 10) in condition 1 and 0.30 kg/m^3/d (COD:N = 20) in condition 2. The MBR was continuously operated for 160 days under the two NLR conditions. Constituents of synthetic wastewater were: glucose (1031 mg/L), NH_4Cl (191 mg/L) and KH_2PO_4 (87 mg/L) as primary nutrients, while $CaCl_2$ (10 mg/L), $MgSO_4\cdot 7H_2O$ (10 mg/L), $FeCl_3$ (3 mg/L), $MnCl_2\cdot 4H_2O$ (2 mg/L) as trace nutrients. $NaHCO_3$ (200 mg/L in condition 1 and 600 mg/L in condition 2). Decrease in pH was observed with change in NLR due to which buffer concentration was increased from 200 to 600 mg/L to adjust the pH between 7 and 8.

2.3. Analytical methods

Parameters such as ammonium nitrogen (NH_4^+–N), nitrite nitrogen (NO_2^-–N), nitrate nitrogen (NO_3^-–N) and phosphate phosphorous (PO_4^{3-}–P), total nitrogen (TN), chemical oxygen demand (COD), mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), and specific oxygen uptake rate (SOUR) were analyzed according to the methods discussed in Khan et al. (2011). Dissolved oxygen and pH in the reactors were measured by DO/pH meter (Oakton PD 300, USA). Sludge particle size distribution was analyzed using a particle size analyzer based on laser scattering principle (LA-300, Horiba, Japan) and the results were reported in percentage volume. Soluble EPS was separated by sludge centrifugation at 5000 rpm for 15 min and analyzing the supernatant. Bound EPS was extraction using cation exchange resin method (Frølund et al., 1996). The soluble or bound EPS was determined as the sum of carbohydrate and protein contents. The phenol–sulfuric method (Dubois et al., 1956) and the Lowry method (Lowry et al., 1951) were used for determination of carbohydrate and protein concentrations, respectively. All samples were measured in duplicates. The membrane fouling in MBR was monitored in terms of trans-membrane pressure (TMP) rise with operational time and fouling resistances at the end of each filtration cycle. TMP was measured continuously with data logging manometer (Super-Scientific 840099, Taiwan). When TMP reached 40 kPa the membrane filtration operation was stopped and membrane was subjected to physical and chemical cleaning as per manufacturer guidelines. Fouling resistances were measured using resistance-in-series model.

3. Results and discussion

3.1. Sludge characteristics

3.1.1. MLSS and MLVSS

MLSS and MLVSS were monitored regularly while maintaining SRT of 30 days. Average MLSS concentrations in MBR were 9.9 ± 1.7 and 10.2 ± 1.8 g/L under condition 1 and 2, respectively while average MLVSS concentrations were 7.6 ± 1.4 and 8.3 ± 1.6 g/L under condition 1 and 2, respectively. The ratio of MLVSS/MLSS in the MBR i.e., 0.76 and 0.81 under conditions 1 and 2, respectively was almost constant during the MBR operation, suggesting that there was no accumulation of inorganic matter and that most of the suspended solids were microorganisms.

3.1.2. Particle size distribution

The particle size distribution was evaluated on the basis of mean particle diameter by its percentage volume in the sample. The average mean particle size of the MBR was 95.52 μm (distribution range of 67–133 μm) in condition 1 and 133.09 μm (distribution range of 3–592 μm) in condition 2 where the average particle size increased while the particle size distribution changed significantly shifting from narrow to broader distribution. Wisniewski et al. (2000) observed the shift towards larger particles with increase in OLR. Similarly, in this study, a significant shift towards wider particle size distribution with increase in NLR was observed depicting that change in NLR can influence the bacterial morphology.

3.1.3. Specific oxygen uptake rate (SOUR)

The SOUR was measured as an indication of microbial activity. The average SOUR values in this study were found to be 76.9 mg O_2/gVSS/h in condition 1 and 68.0 mg O_2/gVSS/h in condition 2. These values suggest that the smaller and relatively evenly distributed particle sizes provided a larger surface area for substrate utilization and oxygen transfer, thus exhibiting higher respirometric activity of microbes and better treatment performance (to be discussed later) under condition 1 as compared to condition 2.

3.1.4. Extra polymeric substance (EPS)

The concentration of soluble EPS which is considered as a major foulant was more in condition 2 (70.18 ± 2.35 mg/L) as compared to condition 1 (54.74 ± 2.18 mg/L). Concentration of bound EPS was found to be 52.53 mg/g VSS in condition 1 and 16.52 mg/g VSS in condition 2. Soluble EPS was predominantly composed of carbohydrate fraction while bound EPS was mainly composed of protein contents. Similar contributions of carbohydrate and protein contents in soluble and bound EPS concentrations were also observed in Khan and Visvanathan (2008) and Sombatsompop et al. (2006). The main reasons for similar EPS trends among these studies can be attributed to similar feed composition, constituent concentrations, and MBR operational conditions. Furthermore, lower EPS concentration under condition 1 as compared to condition 2 could also be one of the reasons for lower membrane fouling. Hence, higher NLR resulted in increase in smaller microbial-floc sizes consequently releasing more soluble EPS into the mixed liquor.

3.2. Treatment performance

The operational performance of MBR was evaluated in terms of organic matter and nutrients removal.

3.2.1. Organic matter removal efficiencies

Average COD concentrations in the effluent were found to be 20.82 ± 12.22 and 30.50 ± 21 mg/L with COD removal efficiencies of 98% and 97% under condition 1 and 2, respectively. Good performance of MBR in removing organic matter and nitrogen has been reported earlier by Wang et al. (2012) which observed 83.3 ± 8.4% COD removal efficiency and 98.1 ± 1.9% NH_4^+–N removal efficiency in treating low strength municipal wastewater. The results indicate that an increase in NLR (decrease in COD:N ratio) did not adversely affect the COD removal efficiency.

3.2.2. Nutrients removal

Nitrogen removal efficiency of MBR was evaluated in terms of NH_4^+–N and TN removal. The average NH_4^+–N removal efficiency was found to be 90.5% in condition 1 and 89.3% in condition 2 while the average effluent concentrations were 4.50 ± 1.50 and 10.52 ± 4.72 mg/L, in condition 1 and 2, respectively. The average TN removal efficiency was 74% in condition 1 and only 35% in condition 2 because NLR was low in condition 1 and most of the influent ammonium nitrogen was removed via microbial assimilation as compared to limited simultaneous nitrification and de-nitrification in condition 2. Influent phosphorous (PO_4^{3-}–P) concentration was maintained at 19.63 ± 0.60 mg/L. The average removal efficiency of PO_4^{3-}–P in MBR was 38% and 14% under condition 1 and 2, respectively. High nitrogen loading rates may have resulted in excessive growth of heterotrophic microorganisms and reduce the growth of phosphorus accumulation microorganisms (PAOs) and slow growing de-nitrifiers.

3.3. Fouling behavior

3.3.1. Membrane filtration performance

In order to investigate the fouling behavior, the change in temporal TMP was monitored at constant flux of 8.75 L/m^2 h (sub-critical flux). The typical TMP profiles for condition 1 and condition 2 are shown in Fig. 2. The TMP variations in MBR in both the conditions are characterized by three step fouling phenomenon i.e., an initial short term rapid rise in TMP (stage I), then gradual and slow rise in TMP (stage II) followed by a rapid rise in TMP (stage III) until it reached 40 kPa. In condition 1, average fouling period was 4.5 days while in condition 2, membrane was fouled after 3 days. Assuming constant MLSS concentration of approximately 10 g/L under both the conditions, the high amount of soluble EPS and scattered particle size distribution under condition 2 can be the two predominant factors causing higher fouling.

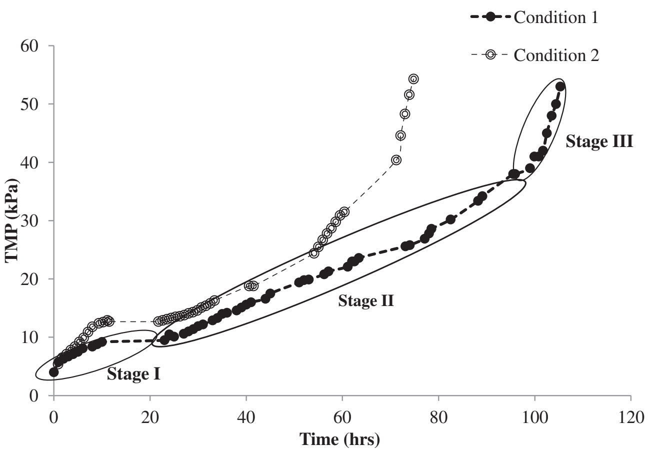

Fig. 2. Trans-membrane pressure (TMP) profiles of the MBR system.

Table 1
Fouling resistances in MBR.

	R_m		R_f		R_c		R_t
	(10^{12}/m)	%	(10^{12}/m)	%	(10^{12}/m)	%	(10^{12}/m)
Condition 1	1.49	11.98	6.61	52.02	4.81	36.35	12.91
Condition 2	1.80	11.10	6.44	38.0	8.93	50.78	17.18

3.3.2. Resistance analysis

The resistance-in-series model was used to evaluate the membrane filtration characteristics whereby the results are reported in Table 1. The total hydraulic resistance (R_t) was found to increase with increasing NLR in condition 2 as compared to condition 1 i.e., 12.91×10^{12}/m in condition 1 and 17.18×10^{12}/m in condition 2. The irreversible fouling resistance (R_f) fraction was almost the same under the two conditions while the cake layer resistance (R_c) was significantly higher in condition 2 as compared to condition 1. This result infers that cake resistance is the major contributor towards membrane fouling in MBR.

4. Conclusions

Submerged MBR exhibited excellent COD removal, good nitrogen removal, but relatively poor phosphorus removal in treating a high strength synthetic wastewater.

Increase in NLR affected the mixed liquor properties especially particle size distribution shifting towards scattered distribution resulting in lower cake layer porosity. Soluble EPS release also increased with higher NLR which adversely affected the fouling propensity. Rapid fouling under condition 2 was mainly attributed to severe cake layer resistance (R_c). This study suggests that the performance of aerobic MBR mostly comprising of heterotrophic bacteria can be appropriate under COD/N ratio of either 20 or above to maintain superior treatment performance and lower membrane fouling.

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