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Removal of Nutrients from Hospital Wastewater by Novel Nano-Filtration Membrane Bioreactor (NF-MBR)

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Abstract: In this paper a pilot-scale membrane bioreactor (MBR) equipped with nano-membranes treating the hospital wastewaters has been studied in order to evaluate the removal of nutrients. The main goal was to examine the feasibility of the NF-MBR used to treat hospital wastewater. Process efficiency was evaluated in terms of COD, NH_3-N, NO_3_N, NO_3_N and PO_4-P removal. The pilot plant located at the Babol Clinic Hospital, Babol, Mazandaran, Iran. The performance of NF-MBR pilot for biological nutrient removal was evaluated during 62 days of operation. The NF-MBR resulting in respective HRT of 2.9 h. The influent COD, NH_3_N and PO_4-P averaged at 688, 11.8 and 3 mg/l respectively. The final treated effluent for the same period averaged 35, 1.69 and 0.89 mg/l respectively. The operating results of the pilot plant demonstrate that the NF-MBR process is both effective and efficient in meeting the water discharge and reuse quality requirements at the hospital. The NF-MBR showed a high COD removal, stable and complete nitrification. The biological nutrient removal efficiencies were high from the start of operation with COD and NH_3-N removal efficiencies of 92 ± 4% and 88 ± 2, respectively. During the course of the experiment PO_4-P removal efficiencies increased and finally PO_4-P removal efficiency of 68% was achieved. Indicating that the NF-MBR effluent can be directly discharged into natural waters and it's suitable for reuse.

Key words: Nano-Filtration Membrane bioreactor (NF-MBR) · Hospital wastewater · Nutrients · Flux · Nitrogen · Phosphorous

INTRODUCTION

Several sources of sewage could contribute to the load of pollutants released in water bodies, particularly hospital sewage [1]. Hospitals are known to be intensive consumers of water, thus generating substantially higher wastewater flows than conventional households (400-1200 l/bed.d vs. 100 l/capita.d) [2, 3]. Hospital wastewater is considered a complex mixture populated with pathogenic microorganisms. Hospital wastewaters contain a variety of toxic or persistent substances such as pharmaceuticals, radionuclide, solvents and disinfectants for medical purposes in a wide range of concentrations due to laboratory and research activities or medicine excretion [4]. The contact of hospital pollutants with aquatic ecosystems leads to a risk directly related to the existence of hazardous substances, which could damage the natural environment and create a biological imbalance [5-7].

Nowadays, nutrient removal has attracted great attention in wastewater treatment for reuse [1]. As the discharge regulation limits of total nitrogen and phosphorous to surface and ground stream become stringent, the removal of them to meet the discharge limits was necessary [1].

Different systems had been used for treatment of hospital wastewater such as activated sludge and ozonation. Submerged hollow fiber membrane bioreactor

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(MBR) was also proposed in treating the hospital wastewater [6, 8, 9]. MBR is the key element in wastewater treatment for reuse and ready to advance water sustainability. The main feature of MBR is a compact treatment technology which has several advantages over conventional biological systems, such as high effluent quality, excellent microbial separation ability, absolute control of sludge and hydraulic retention times (SRTs and HRTs), high biomass content, low-rate biomass production, small footprint and flexibility in operation [2].

Many researchers have focused on the removal of nutrients by various treatment configurations. Maranon reported TN removal of 80%, while their COD removal was over 90% using a complex biological treatment combining anoxic-anaerobic-aerobic processes [10]. In another investigation was obtained partial nitrification to nitrite using membrane bioreactor systems obtained in an efficiency of 65.5% for the operating conditions at HRT of 18 h and MLSS of 7650 mg/l [11]. Lin [12] reported that the removal of NH_4 and COD had efficiencies of 75 and 92% respectively, in a moving-fixed bed biofilm reactor with aerobic and anoxic phases that give a limited nitrification and COD oxidation.

In particular, there is a growing interest in using low-pressure membrane bioreactor (MBR) coupled with microfiltration (MF) or ultrafiltration (UF) for simultaneous organic and nutrient removal but there was no investigation for employment of nanofiltration in the MBR.

The objective of this study was to evaluate the efficiency of hospital wastewater treated by a combination of biological degradation and nano-membrane process. The use of this combined system would ensure that organic matter would not be released to the environment through hospital wastewater discharge.

MATERIALS AND METHODS

The hospital wastewater stream for continues operation of MBR pilot plant were obtained from a general hospital occupying a conventional activated sludge treatment plant (CASP). Analyses were carried out on composite mean samples from influent, effluent and the mixed liquor. The analyses were made according to standard methods procedures [13].

The pilot plant is located at the Babol Clinic Wastewater Treatment Plant (WWTP), Babol, Mazandaran, Iran. The pilot plant treats Hospital wastewater with a ratio of nutrients (C: N: P) of 100:8:0.4. The aim of this study was to demonstrate the complementarities of combining membrane bioreactor (MBR) treatment with nanofiltration (NF) membrane filtration for the removal of nutrients from hospital wastewater for potential reuse of water.

A laboratory scale NF-MBR system was employed in this study (Figure 1). The raw wastewater is collected directly from the aeration`s tank inlet, after passing a screen. The wastewater is pumped to the pilot plant using a centrifuge pump, crossing fine screen with a 1 mm pore size (to prevent large solids from entering the bioreactor and damaging the nano-membranes) and stored in a 300 L equalization tank. The main function of the equalization tank is to adjust the flow and quality of wastewater. This consisted of a plexiglas reactor with an active volume of 80 l, an air pump, a pressure sensor and influent and effluent pumps. Two submerged hollow fiber nano filtration membrane modules were used in this apparatus which operated at a constant value of the net filtration flow rate (0.5-0.8 m^2/day). The selected flow allows obtaining in the pilot plant an hydraulic retention time (HRT) of 2.9 h.

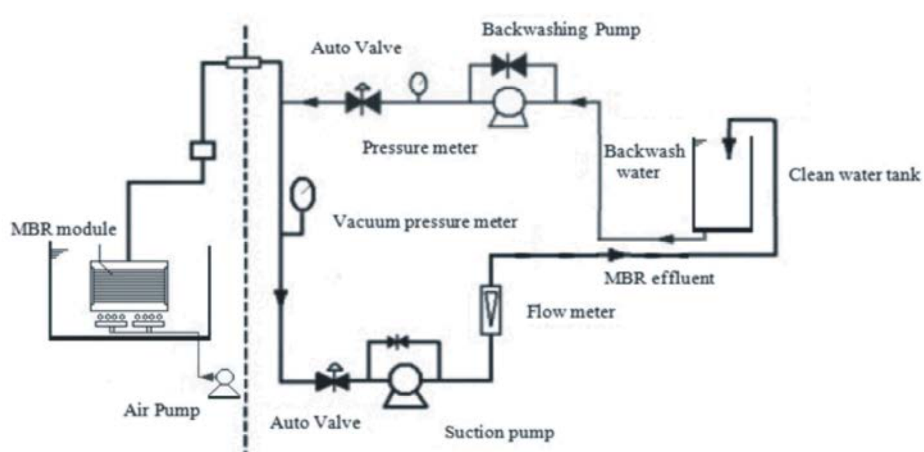

Fig. 1: NF-MBR pilot plant schematic

This membrane has a nominal pore size of 40 nm. Each module had an effective membrane surface area of 4 m^2. An air pump with a maximum air flow rate of 450 l/min was used to aerate the NF-MBR system for biological growth and also used to provide a constant air flow rate through the membrane module to reduce fouling and cake formation via a diffuser located at the bottom of the reactor. The permeates is obtained by applying a vacuum pressure drop over the membranes using a second positive advance pump. Permeate is separated in a different tank (backwash and permeate tank), to ensure constantly sufficient water for backwashing the membranes. Ultimately, the treated effluent collected in the permeate tank is discharged from the pilot plant to the WWTP sewer. No biomass was wasted from the NF-MBR during the experiment.

RESULTS AND DISCUSSION

COD Removal Performance: The MBR influent COD varies in a range of 635-744 mg/l in the period of experiments, corresponding to COD loading rates of 38.1-44.67 kgCOD/day. Figure 2 shows the COD concentration of the influent, effluent during the experimental period and COD removal efficiency of the whole system. The influent characteristics were almost the same due to same daily routine work and performance of equalization tank.

Figure 2 indicates that the system attained steady-state effluent as evident from the low standard deviations, which is testament towards the stability of the system. From Figure 2 it can also be seen that the system can provide a consistently high COD removal efficiency. The effluent COD after 20 days was always lower than 50 mg/l. the effluent COD concentration fulfilled the set discharge limits of 50 mg/l and the average removal efficiency was 94 ± 2% and at the end of the run time COD concentration was 35 mg/l. The results also showed that membrane separation played an important role providing the excellent and stable effluent quality due to the pore size and formation of gel layer in.

N-NH_3 and N-NO_2 and N-NO_3 Removal Performance: During the start-up period the NH_3^--N removal efficiency increased rapidly, whereas at day 7, NH_3^--N removal already exceeded 82%. Nitrification onset was rapid, whereas the effluent NH_3^--N concentration was consistently below 3 mg and the average of below 1.69 mg/l which was realized by the good sludge seeding.

The overall NH_3^--N removal efficiency, amounted to 88 ± 2%. Analogue to the effluent quality for removal of organic matter, NH_3^--N removal in the pilot plant NF-MBR was constantly high, resulting in an effluent quality with little variation, despite of influent variability. The influent NH_3^--N varied from 10.1 to 14 mg/l with the average of 11.8 mg/l.

The variations of NH_3^--N concentration in the influent and membrane effluent, as well as NH_3^--N removal by the bioreactor and by the whole system are presented in Figure 3.

Figures 4 and 5 show the biological nitrogen removal (BNR) during the entire experimental run. NO_2^--N removal efficiencies was high during the run period (after 20 days, it always was more than 80%). In general, nitrification rate was high throughout the experimental runs achieving 1 mg/l NH_3^--N in the effluent.

The NO_2-N concentrations of the supernatant and effluent were not approximately same, which could be explained by the fact that the NO_2-N was ion which could not passed entirely through the nanopores of the membrane. As a result, the nitrite nitrogen was removed by two mechanisms, first the microorganisms in the MBR and then by nano hollow fiber membranes.

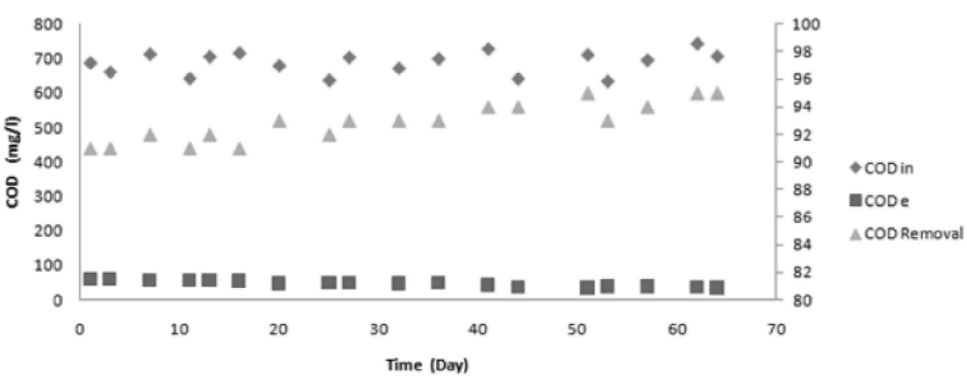

Fig. 2: COD concentration and removal performance

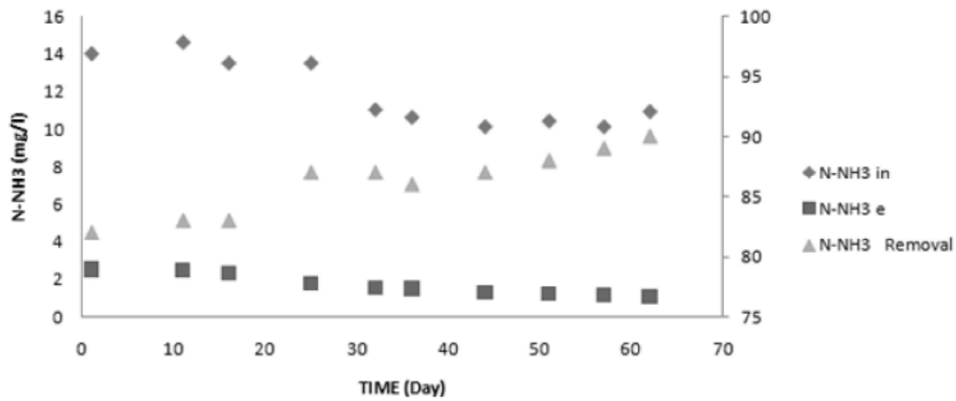

Fig. 3: NH_3^--N concentration and removal performance

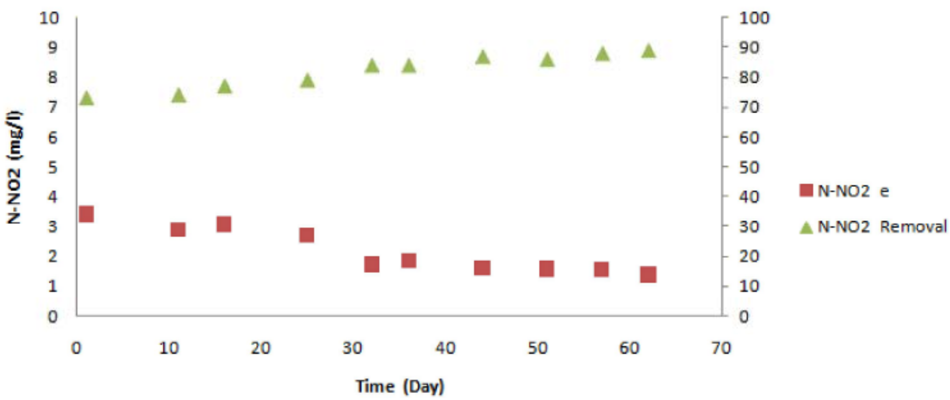

Fig. 4: NO_2^--N concentration and removal performance

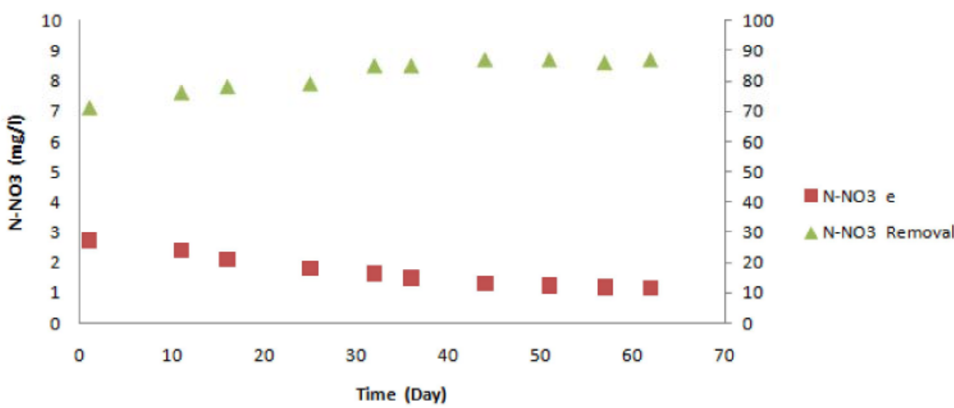

Fig. 5: NO_3^--N concentration and removal performance

The high membrane interception of the MBR provides the suitable growing and regenerating conditions for the long sludge age required for nitrobacteria.

The wastewater composition heavily affects the nitrification process: the presence of inhibiting compounds, common in hospital wastewater [14, 15], did not allow obtaining a complete nitrification despite the high sludge age. This finding may depend on many factors such as: (i) the different micro-organism selection; (ii) the higher bio-availability of substrates which can be due to the smaller size of flocs in MBR sludge; and (iii) the tendency of nitrifiers to grow inside, rather than on the surface, of the flocs [15-17].

The variation of influent and effluent NO_3^--N concentration with time during the experiment is illustrated in Figure 5 indicates the NO_3^--N removal by the NF-MBR pilot plant. The influent NO_3^--N fluctuated from 9.06 to 11.03 mg/l but the effluent NO_3^--N was always under 2.5 mg/l and at final day of experiment it was about 1 mg/l and the removal of NO_3^--N was 87%. NO_3^--N removal efficiency of the NF-MBR had an adequate stability with time. Also efficiency was increased with a mild slope over the time. The formation of MLSS in the aeration tank and formation of the cake layer on the nano membranes during the experiment was the main reasons for increase of NO_3^--N removal efficiency.

Nitrogen could be removed only by assimilation into biomass or by nitrification-denitrification. The mean ammonia nitrogen concentration in the influent of MBR was in the range of 10-14 mg/l. As nitrifying bacteria had longer generation times, they needed more time to establish and reach sufficient concentrations to nitrify the ammonium. It took another several days until nitrite was completely converted to nitrate. One week later, the ammonium nitrogen concentration in the effluent declined rapidly to 3.5 mg/l and 90% of the ammonia nitrogen was eliminated. Generally, in the MBR system higher sludge concentration limited the oxygen mass transfer through the mixed liquid, consequently caused lower oxygen concentration in some area to create local anaerobic area beneficial for denitrification [18].

In the absence of an anoxic compartment, the denitrification capacity of the MBR system used in this study was limited. Therefore, the high initial TN removal observed here could possibly be attributed to the conversion of dissolved organic nitrogen to biomass, which would then be retained by the membrane [19].

PO_4^{3-}-P Removal Performance: Towards the end of the start-up phase the effluent phosphate concentrations were lower than 2 mg/l PO_4^{3-}-P, set as discharge limit in Iran legislation. Figure 6 shows that the effluent phosphate concentrations never exceeded the set limits during the experiments. The highest effluent concentration detected in this period was around 1.5 mg PO_4^{3-}P. At the end of the period of run the P-removal efficiency improved from 46 to 68%.

The high nitrate concentration in the reactor (11 mg/l NO_3^--N) at the beginning also provoked a limitation of the P removal [20]. Nevertheless, at the end of experimental period, the P concentration decreased significantly, achieving an effluent concentration lower than 0.9 mg PO_4^{3-}-P.

To obtain higher removal percentages of P, anaerobic conditions are required for the uptake and storage of easily biodegradable organic matter and also P release, to finally accumulate phosphate under anoxic or aerobic conditions [20 -22].

Although still little is known about the microbiology of phosphorous removal, the deterioration of P removal in processes has been linked to e.g. the presence of nitrate in the anaerobic stage [23], potassium and/or magnesium limitation to stabilize the strong negative charge of poly-P [24, 25], over-aeration [26] and the microbial competition of glycogen accumulating organisms (GAOs) with poly-phosphate accumulating organisms (PAOs) [20, 27].

MLSS and DO Concentration: During the 20 days start-up period, MLSS concentration increased from 500 mg/l to 1027 mg/l. Figure 7 shows the time-dependent variations of sludge concentration in the bioreactor during the experimental study. During the first 30 days, the sludge concentration increased so fast because of good seeding and microorganism acclimatization. No sludge was withdrawn from the membrane bioreactor during 62 days operation except for MLSS concentration examination, which resulted in a long SRT. This long SRT after 55 days allowed the excess sludge being oxidized in the bioreactor, which kept the sludge concentration constant. The MLSS concentration increased from 1027 to 6078 mg/l in the aeration tank.

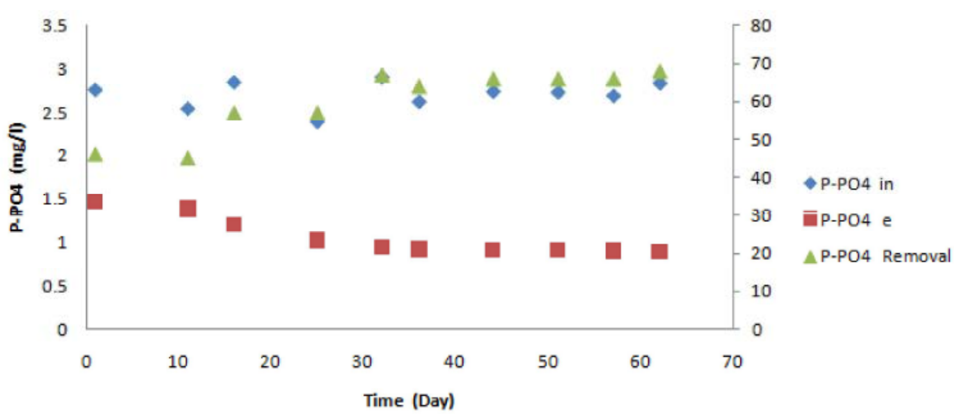

Fig. 6: PO_4^{3-}-P concentration and removal performance

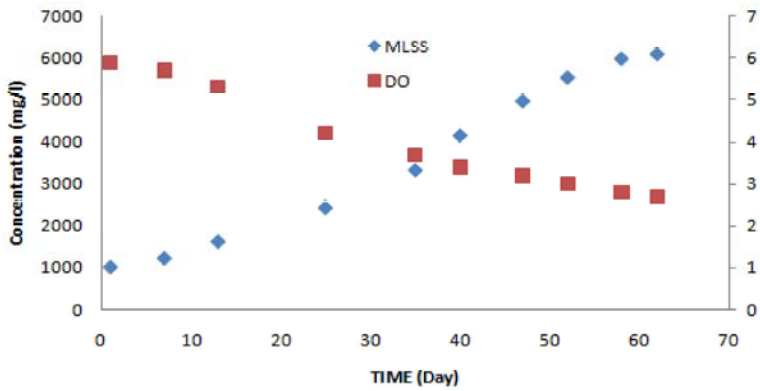

Fig. 7: MLSS and DO concentration in bioreactor

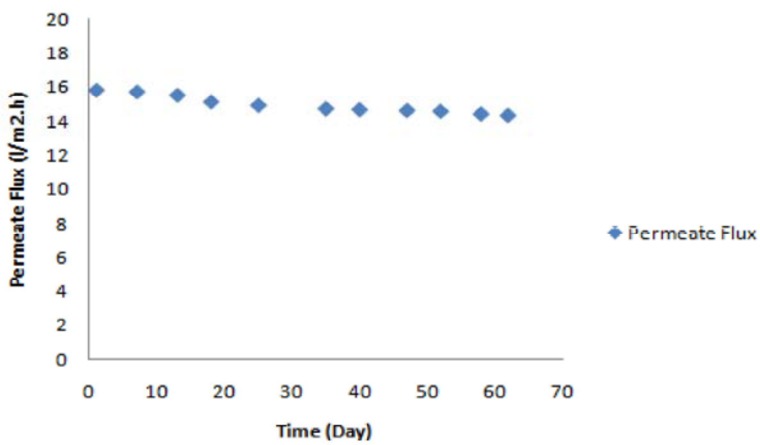

Fig. 8: Variation of the membrane permeate flux as a function of filtration time

In addition, DO in MBR system was determined during the operation shown in Figure 7. It can be seen that DO in MBR decreased as the operating time increased from 5.9 mg/l of the first day to 2.7 mg/l at the end of operation.

Flux: The rejection of contaminants ultimately places a fundamental constraint on all membrane processes. The rejected constituents in the retentate tend to accumulate at the membrane surface, producing various phenomena which lead to a reduction in the flow of water through the membrane (i.e. the flux) at a given transmembrane pressure (TMP), or conversely an increase in the TMP for a given flux (reducing the permeability, which is the ratio of flux to TMP). Even under very low fluxes, a slight increase of TMP can be observed [28].

The start-up of the NF-MBR system was investigated step by step. First, during the adjustment period the activated sludge was acclimated to the hospital wastewater and then the membrane bioreactor was operated by almost constant flux. Figure 8 shows the exact flux variations during the operation. The TMP increases gradually at an acceptable rate, such that chemical cleaning was not necessary. The pressure increase was caused by the thickening of the gel layer on the membrane surface. However, the membrane was not chemically cleaned during the 2 months' operation. A 5-10 min/day backwash program was implied to recover the permeability and cleaning the nano-membrane surface in this experiment.

Figure 8 shows the evolution of the membrane permeate flux as a function of filtration time. After 15 days a gel layer including polysaccharides and protein was formed on the membrane surface and slightly decreased the flux. In fact, the nano-membrane in MBR did not show any Considerable permeate flux decline over approximately 62 days of filtration time.

CONCLUSION

The system operated steadily for 62 days, which demonstrated that the start-up and operation strategies for the pilot were successful. The system is simple in operation and cleaning and no need for chemical cleaning to recover the flux rate. Flux rate of NF-MBR was equal to 16 $l/m^2.h$. A hydraulic retention time in the bioreactor of 2.9 h. was sufficient to obtain degradation of nutrients such as nitrogen and phosphorus when operating at a sludge concentration of 6000 mg/l.

Under optimum conditions, maximum reduction of COD, NH_3-N, NO_2-N, NO_3-N and PO_4-P using NF-MBR at the final days of experiment was achieved. The treatment performance of NF-MBR system was very stable and on a high level. The research proves that the process of eliminating pollutants from hospital wastewater proceeds effectively and removal in 95, 90, 89, 87 and over 68% for COD, NH_3-N, NO_2-N, NO_3-N and PO_4-P were obtained respectively. It is clear that nutrients with smaller molecular size appeared in the final permeate more often and at higher concentrations.

The majority of compounds that were in effluent are hydrophilic, because most hydrophobic compounds have been effectively removed by the MBR treatment process. Adsorption of these hydrophilic compounds to the membrane is not expected to be significant. Furthermore, hydrophobic compounds appear to be more permeable through these nano-membranes than their hydrophilic counterparts.

Results reported here confirm the capacity of NF-MBR to act as effective barriers against nutrients in the hospital wastewater. These results also highlight the merit of coupling nano-membrane filtration with MBR treatment for complementary removal of nutrients such as nitrogen and phosphorus.

REFERENCES

1. Tatiana, P., M. Dalton, C. Guilayn, L. Rose, A. Maria, P. Miagostovich, 2011. Quantification and molecular characterization of enteric viruses detected in effluents from two hospital wastewater treatment plants. Water Research, 45: 1287-1297.
2. Gautam, A.K., S. Kumar and P. Sabumon, 2007. Preliminary study of physicochemical treatment options for hospital wastewater. Journal of Environmental Management, 83(3): 298-306.
3. Suarez, S., J. Lema and F. Omil, 2009. Pre-treatment of hospital wastewater by coagulation-flocculation and flotation. Bioresource Technology, 100: 2138-2146.
4. Verlicchi, P., A. Galletti, M. Petrovic and D. Barcelo, 2010. Hospital effluents as a source of emerging pollutants: An overview of micropollutants and sustainable treatment options. Journal of Hydrology, 389: 416-428.
5. Emmanuel, E., Y. Perrodin, G. Keck, M. Blanchard, P. Vermande, 2005. Ecotoxicological risk assessment of hospital wastewater: a proposed framework for raw effluents discharging into urban sewer network. Journal of Hazardous Materials, A117: 1-11.
6. Kajitvichyanukul, P. and N. Suntronvipart, 2006. Evaluation of biodegradability and oxidation degree of hospital wastewater using photo-Fenton process as the pretreatment method. Journal of Hazardous Materials, B, 138: 384-391.
7. Kajitvichyanukul, P., M. Lu, C. Liao, W. Wirojanagud and T. Koottatep, 2006. Degradation and detoxification of formaline wastewater by advanced oxidation processes. Journal of Hazardous Materials, 135: 337-343.
8. Qiaoling, L., Z. Yufen, C. Lingyun and Z. Xiang, 2010. Application of MBR for hospital wastewater treatment in China. Desalination, 250: 605-608.
9. Xianghua, W., D. Hangjiu, H. Xia and L. Ruopeng, 2004. Treatment of hospital wastewater using a submerged membrane bioreactor. Process Biochemistry, 39: 1427-1431.
10. Maranon, E., L. Castrillon, L. Garcia, I. Vazquez and Y. Fernandez-Nava, 2008. Threestep biological process for the treatment of the liquid fraction of cattle manure. Bioresource Technol., 99: 7750-7757.
11. Francisco, A., R. Reboleiro-Rivas and J. González-López, 2012. Comparative study of the use of pure oxygen and air in the nitrification of a MBR system used for wastewater treatment. Bioresource Technology, 121: 205-211.
12. Lin, Y., 2008. Kinetics of nitrogen and carbon removal in a moving-fixed bed biofilm reactor. Applied Mathematical Modelling, 32: 2360-2377.
13. APHA, 2005. Standard methods for the examination of water and wastewater. 21st ed, American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC. USA.
14. Klinkow, N., J.Oleksi-Frenzel and M. Jekel, 1998. Toxicity-directed fractionation of organic compounds in tannery wastewater with regard to their molecular weight and polarity. Water Res., 32(9): 2588-2592.
15. Giulio, M., M. Gualtiero, L. Salvadori, B. Claudia and L. Claudio, 2008. Process efficiency and microbial monitoring in mbr (membrane bioreactor) and casp (conventional activated sludge process) treatment of tannery wastewater. Bioresource Technology, 99: 8559-8564.

16. Luxmy, B., F. Nakajima and K. Yamamoto, 2000. Analysis of bacterial community in membrane-separation bioreactors by fluorescent in situ hybridization (FISH) and denaturing gradient gel electrophoresis (DGGE) techniques. Water Sci. Tech., 41(10-11): 259-268.
17. Sofia, A., W. Liu, S. Ong and W. Ng, 2004. In situ characterization of microbial community in an A/O submerged membrane bioreactor with nitrogen removal. Water Sci. Technol., 50(8): 41-48.
18. Xiufen, L., F. Gaoa, Z. Hua, G. Dua and J. Chen, 2005. Treatment of synthetic wastewater by a novel MBR with granular sludge developed for controlling membrane fouling. Separation and Purification Technology, 46: 19-25.
19. Abdulhakeem, A., N. Tadkaew, A. McDonald, S. Khan, W. Price and L. Nghiem, 2010. Combining MBR and NF/RO membrane filtration for the removal of trace organics in indirect potable water reuse applications. Journal of Membrane Sci., 365: 206-215.
20. Monclús, H., J. Sipma, G. Ferrero, J. Comas and I. Rodriguez-Roda, 2010. Optimization of biological nutrient removal in a pilot plant UCT-MBR treating municipal wastewater during start-up. Desalination, 250: 592-597.
21. Puig, S., M. Coma, H. Monclús, M. Van Loosdrecht, J. Colprim and M. Balaguer, 2008. Selection between alcohols and volatile fatty acids as external carbon sources for EBPR, Water Res., 42(3): 557-566.
22. Mino, T., M. Van Loosdrecht and J. Heijnen, 1998. Microbiology and biochemistry of the enhanced biological phosphate removal process, Water Res., 32(11): 3193-3207.
23. Kuba, T., A. Wachtmeister, M. Van Loosdrecht and J. Heijnen, 1994. Effect of nitrate on phosphorus release in biological phosphorus removal systems. Water Sci. Technol., 30(6): 263-269.
24. Brdjanovic, D., C. Hooijmans, M. Van Loosdrecht, G. Alaerts and J. Heijnen, 1996. The dynamic effects of potassium limitation on biological phosphorus removal. Water Res., 30(10): 2323-2328.
25. Pattarkine, V. and C. Randall, 1999. The requirement of metal cations for enhanced biological phosphorus removal by activated sludge. Water Sci. Technol., 40(2): 159-165.
26. Brdjanovic, D., A. Slamet, M. Van Loosdrecht, C. Hooijmans, G. Alaerts and J. Heijnen, 1998. Impact of excessive aeration on biological phosphorus removal from wastewater. Water Res., 32(1): 200-208.
27. Thomas, M., P. Wright, L. Blackall, V. Urbain and J. Keller, 2003. Optimisation of Noosa BNR plant to improve performance and reduce operating costs. Water Sci. Technol., 47(12): 141-148.
28. Judd, S., 2006. The MBR book principles and applications of membrane bio-reactors in water and wastewater treatment. Elsevier, Grate Britain (London).