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Industrial wastewater treatment by using of membrane

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Abstract. In this work, treatment of real hypersaline refinery wastewater by hollow fiber membrane bioreactor coupled with reverse osmosis unit was studied. The ability of HF-MBR and RO developed in this work, was evaluated through examination of the effluent properties under various operating conditions including hydraulic retention time and flux. Arak refinery wastewater was employed as influent of the bioreactor which consists of an immersed ultrafiltation membrane. The HF-MBR/RO was run for 6 months. Average elimination performance of chemical oxygen demand, biological oxygen demand, total suspended solids, volatile suspended solids, total dissolved soild and turbidity were obtained 82%, 89%, 98%, 99%, 99% and 98% respectively. Highly removal performance of oily contaminant, TDS and the complete retention of suspends solids implies good potential of the HF-MBR/RO system for wastewater refinement.

Keywords: Hollow Fiber Membrane Bioreactor (HF-MBR); refinery wastewater; wastewater treatment; BOD_5; COD; RO

1. Introduction

The most important waste from gas and oil industry is undoubtedly produced water that is produced as a byproduct along with the oil and gas. The generation of produced water all over the world is estimated about 250 milion barrels per day. The produced water contains a wide variety of organic and inorganic compound. For instance, its TOC concentration could be up to 1500 mg/L and its salt concentration varies from 1 to 300,000 mg/L (Fakhru'l-Razi *et al*. 2009, 2010).

Increasing volume of demanded freshwater from one side and wastewater of oil industry from the other side make scientists try to find a suitable method for treatment of wastewater because water pollution issue shoud be solved, particularly in water-stressed regions (Fakhru'l-Razi *et al*. 2009). Photo-electro catalytic refinement, hydro cyclones, coagulation, and flocculation are some of physical and chemical procedures that can be employed to decrease hydrocarbon contaminants (Fakhru'l-Razi *et al*. 2009). Considering the fact that the mentioned methods are just suitable for primary treatment; the wastewater should be treated by more efficient operations before discharge to environment or further reuses (Campos *et al*. 2003, Pendashteh *et al*. 2012).

An affordable and environmental friendly method is biological refinement of wastewater. However it is noteworthy that metabolism of microorganisms within the activated sludge may be affected by salinity in hypersaline wastewater. Therefore halophilic microorganism was employed

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in biological treatment of saline wastewater (Tellez *et al*. 2002, Zhao *et al*. 2006). In the other hand, membranes due to their unique properties have used in some applications worldwide such as gas separation, waste water treatment and etc (Rácz *et al*. 2015, Tahvildari *et al*. 2015, Saha *et al*. 2014, Fazaeli *et al*. 2015, Razavi *et al*. 2015a, b, Kertész 2014, Melin *et al*. 2006, Clara *et al*. 2005, Miramini *et al*. 2013, Razavi *et al*. 2013, Rezakazemi *et al*. 2013, Gryta *et al*. 2006, Ghadiri *et al*. 2013). More efficient treatment can be attained by use of membrane separation technique and activated sludge together, which have more advantages such as ability of running the unit from high mixed liquor suspended solids (MLSS) concentration, minimal footprint, beneficial disinfection capability, higher volumetric loading and high effluent quality (Le-Clech *et al*. 2006, Shariati *et al*. 2013). MBR methods that use the micro or ultrafiltration have advantages of complete retention of bacterial flocs and nearly whole of suspended solids in the bioreactor (Le-Clech *et al*. 2006). According to the literature, other membranes have been used for treatment of oily wastewater (Wang *et al*. 1998, Kong and Li 1999, Lee *et al*. 1994, Andrzej and Field 1996, Scholz and Fuchs 2000). Flexibility and easier operation and ability of handling different feed quality make the MBR method to be a favorable biological method (Venkata Mohan *et al*. 2005). High salt concentrations results to weak settle ability and therefore results the effluent become turbid (Kang *et al*. 2003). Both of municipal and industrial wastewater has been treated by MBR widely (Chiemchaisri and Yamamoto 1994, Shim *et al*. 2002, Lau and Ismail 2009, Grélot *et al*. 2009, Yuliwati and Ismail 2011).

Microfiltration (MF), ultrafiltration (UF) and reverse osmosis (RO) are some of dual-membrane methods that have increasing attraction because of good performance and easy and affordable operation (Reith and Birkenhead 1998, del Pino and Durham 1999). In this process, suspended solids and colloidal components are eliminated by MF or UF, and then dissolved solids, organic and ionic components are eliminated by the RO. According to this point that MBR enables to secondary treatment of wastewater and also pretreatment for RO, MBR/RO can be considered as a suitable method for refinement of raw sewage (Tam *et al*. 2007).

In previous studies, ability of the MBR/RO was investigated just for a synthetic wastewater which doesn't contain of pollutants as much as a real wastewater. It is obvious that results of such studies will not directly use for the real wastewater with a wide variety of organic and inorganic components. Authors could find only some publication in which an aerobic HF-MBR joined with RO unit for treatment of actual wastewater was investigated, so this present study develops a HF-MBR/RO system and evaluates its ability for treatment of the actual high TDS oil refinery wastewater.

2. Materials and methods

2.1 Explanation of the pilot plants

A bench-scale system consist of a submerged HF-MBR with a tank of activated sludge which coupled with RO unit, was developed in the Environmental Engineering research laboratory of Arak University and used for our experiments. Fig. 1 shows the schematic plan of the HF-MBR/RO. Polypropylene and polyamide membranes were used as the hollow fiber membrane and RO membrane, respectively. Polymeric membrane was selected because of the membrane surface is more than ceramic membrane. The average pore size of hollow fibers was 0.1 to 0.2 micrometer (Pishtaz Polymer Sepahan Co., Iran). The membrane had a effective filtering region of

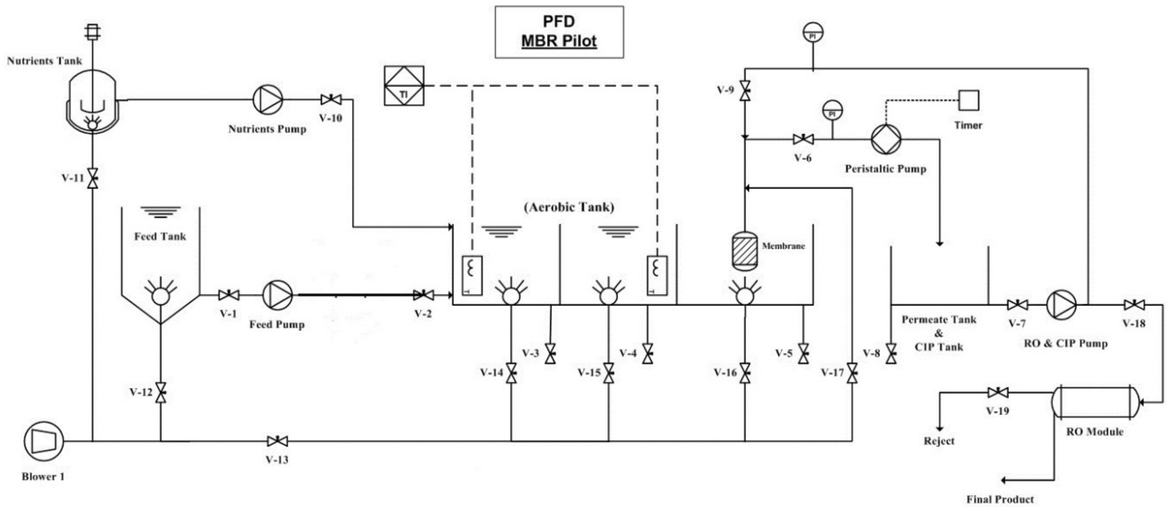


Fig. 1 Schematic diagram of the HF-MBR/RO system

Table 1 Hollow fiber membrane module characteristics

	Parameter	Value	Unit
1	Effective length	16	cm
2	Internal diameter	0.25	mm
3	Outer diameter	0.4	mm
4	Number of fibers	1940	-
5	Pore size	0.1-0.2	μm
6	Filtration area	0.39	m^2
7	Membrane material	PP	-

0.39 m^2. Other characterizations of membrane were shows in Table 1. Also the image of some part of system demonstrated in Fig. 2 (consist of 2a: blower system, 2b: peristaltic pump and RO module, 2c: hollow fiber membrane in bioreactor tank, 2d: hollow fiber membrane after a few weeks of work).

A peristaltic pump (Etatron D.S., Italy) with an Intelli-Cycle timer without back-flush was used for filtration method. Both of the MBR sludge and wastewater supplied from the Arak Oil Refining Co., Iran. The activated slidge employed in these studied was adapted within the submerged MBR rig. The raw wastewater used in this work has COD of 580 ppm, BOD of 203 ppm, TDS of 2100 ppm, pH of 7.6 and turbidity of 40 NTU. Table 2 indicates the major operation parameters. A dispersed air-flow rate of 70 lit/min was provided by aeration apparatus located under the membrane modules. A constant temperature of 20°C was selected for the system. A constant biomass volume was obtained by a feed pump which was equipped with a level control. The HRT of reactor varied from 25 to 36 h and mixed liquor suspended solid (MLSS) concentration varied from 3 to 6.6 g/L. This concentration was maintained at this amount through the partially taking off the sludge from the bioreactor. Dissolved oxygen amount was kept at a desirable level through the additional air diffuser to provide a higher aeration. Fresh water was

(a) (b) (c) (d)

Fig. 2 Image of some part of system

Table 2 Operation conditions and measurements during different Hydraulic Retention Time

Parameter \ Operation time	1-25 days	25-45 days	45-60 days
Hydraulic retention time (hr)	36	30	25
Flux (L/ m^2.hr)	0.846	1.025	1.205
Q_{out} (L/hr)	0.33	0.4	0.47

flushed to the membrane system for physical cleaning. Although not any type of chemical cleaning was applied during the whole research.

2.2 Analytical methods

Disolved oxygen (DO), pH, temperature, MLVSS, COD and BOD_5 were checked to evaluate the avtivated sludge bioactivity. The COD, color and turbidity, within the feed and also within the penetrate, were measured every week. All of the aforementioned parameters were determined regard to the standard technique (APHA 2005).

3. Results and discussion

3.1 Overall performance

Performance of the MBR and also RO unit within the pilot plan were explored frequently via evaluate of their operation and quality of treated wastewater.

3.1.1 MBR unit efficiency

The MBR was employed for eliminate the organic compound and nitrogen from the wastewater and also pretreatment for RO unit. The results show that the MBR performance was stable and effective.

3.1.1.1 Sludge adaptation and characteristics

With use of a fed-batch system, the sludge inoculant was grown up during the whole of adaption period. As it can be seen in Fig. 3, the MLSS concentration increased from 3 to 6.6 g/lit after 8 weeks. Actual refinery wastewater was used as the feed of the MBR. Therefore the feed concentration increased because the MLSS concentration was increased. After the fed-batch operation was finished, the MLSS concentration reached to 6.6 g/lit and was kept constant for the subsequent continuous process. The removal performance of COD varied from 55% to 75% during whole of the fed-batch process. Fig. 4 demonstrates the COD_{in} and COD_{out} concentrations during entire the adaption time.

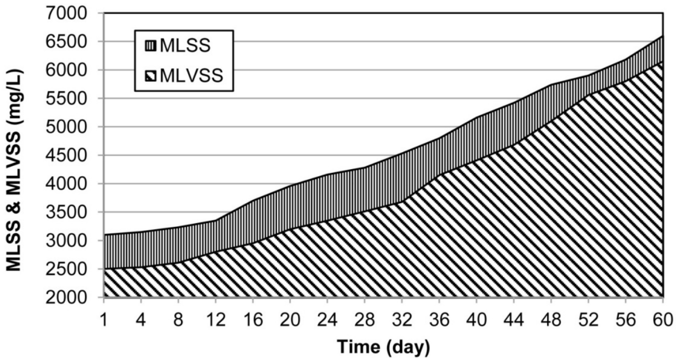


Fig. 3 Profile of MLSS, MLVSS during the adaption period

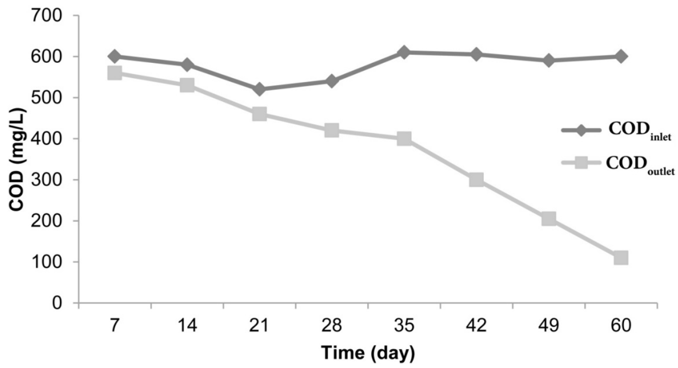


Fig. 4 Profile of $COD_{in,}$ and COD_{out} during the adaption period

3.1.1.2 MLSS, MLVSS, VSS, and TSS concentration

As it can be seen in Fig. 3, MLSS and MLVSS concentrations increased from minimum concentration to about 6.6 and 6.1 g/lit respectively after adaptation period of 2 months. TSS and VSS concentrations in effluent were considerably low and removal performance of VSS and TSS under all operating conditions was over than 98% (see Fig. 5). This is due to the UF membrane had been employed here, which low levels of TSS and VSS are in accordance with the previous investigation (Fazeli *et al*. 2012).

3.1.1.3 Turbidity and SS within the effluent

During the entire process, the SS concentration within the effluent was about 1-2 %. Therefore the effluent only contains of dissolvable pollutants. Fig. 6 reveals that elimination performance of SS was 98-99%. Therefore it can be concluded that the membrane is very appropriate for such separation. Turbidity of the raw sewage varied from 35 to 45 NTU. The filtrate turbidity was usually less than 1 NTU because of efficient separation ability of the membrane. This result is in agreement with Pendashte *et al*. work (Pendashteh *et al*. 2012). Pictures of treated and raw wastewater are shown in Fig. 7. Treated wastewater with a minimum turbidity reveals excellent elimination performance, compared with raw wastewater.

3.1.1.4 The COD and BOD5 removal within the HF-MBR procedure

The results showed that in the habituation period, removal performance of COD and BOD were

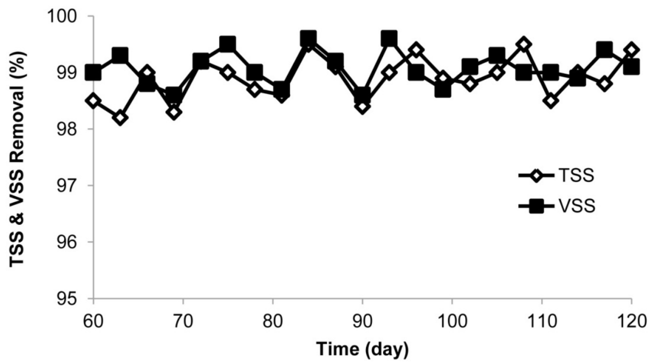


Fig. 5 TSS and VSS concentrations at inlet and outlet flow

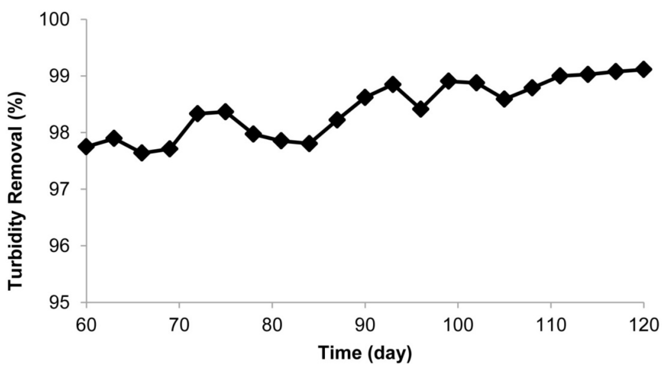


Fig. 6 Turbidity and SS within the effluent

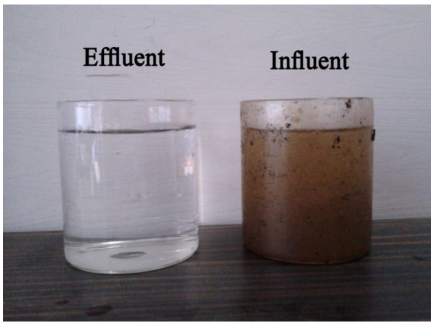
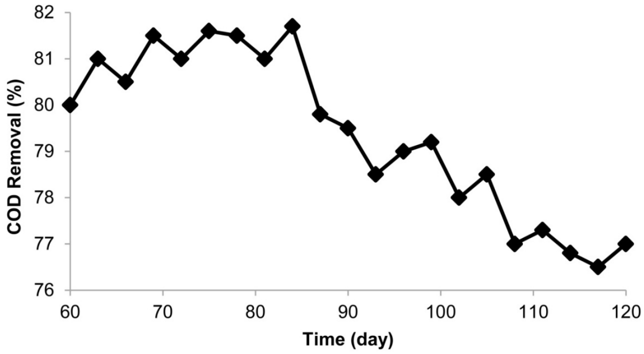
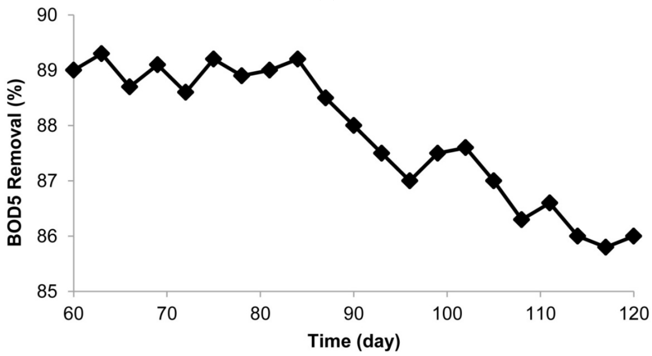


Fig. 7 Turbidity (visual) comparison of treated wastewater and raw wastewater



(a)



(b)

Fig. 8 (a) The COD during the operation time; (b) The BOD_5 during the operation time

at the minimum amount due to this fact that biomass rise was not adequate in the process. Variation of COD and BOD removal efficiency are shown in Figs. 8(a) and 8(b), respectively. As it can be observed in the Fig. 8, operation time of 60 to 8 days results in the best COD and BOD elimination performance. Also it can be find that the COD and BOD concentrations in the effluent were at a significantly low amount.

Various HRTs of the system and effect of at period duration of 24 h during the entire experiment can be seen in Table 3. According to the table 3, as HRT reduces from 36 h to 30 h and then to 20 h, the COD elimination reduces from 81.08% to 78.92% and then to 78.92% and also

Table 3 COD and BOD_5 removal during different Hydraulic Retention Time

Parameter \ Operation time	1-25 days	25-45 days	45-60 days
Hydraulic retention time (hr)	36	30	25
BOD Removal (%)	89	87.6	86.1
COD Removal (%)	81	79	77

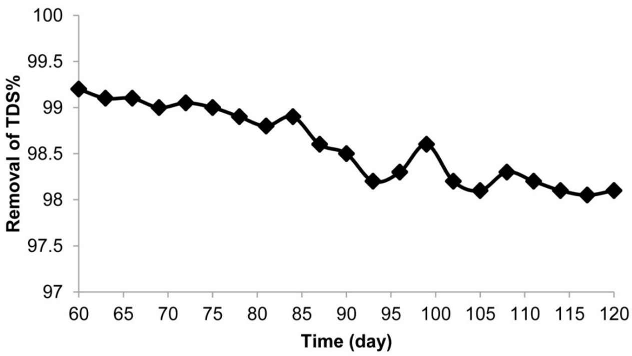


Fig. 9 Rejection efficiency of TDS by the RO membrane during the operation time

BOD elimination reduces from 89% to 87.58% and then to 86.14%. Minimum concentrations of COD and BOD in the effluent were obtained at HRT of 36 h. It should be noted that the effluent of HF-MBR had a low ratio of BOD/COD (0.18-0.22). The following low ratio perhaps probably quite possibly is caused by presence of there bellious organic elements in the crude oil (Neff 2002) or microorganisms disolveable metabolic byproducts, that can hold throughout the membrane (Pendashteh *et al*. 2012, Scholz and Fuchs 2000).

3.1.2 RO performance

Permeate pollutant concentration and the membrane rejections were applied to investigation of the RO performance. As it can be seen in Eq. (1), RO rejection can be calculated based on conductivity (Tam *et al*. 2007)

$$\text{Rejection (\%)} = \left[1 - \frac{\text{Permeate conductivity}}{\text{(Feed water conductivity + Concentrate conductivity)/2}}\right] \times 100 \qquad (1)$$

Regard to the obtained results, the concentration of the pollutants were considerably low and also quality of the RO penetrate was excellent. Fig. 9 shows the ability of the RO membrane to rejection of TDS based on conductivity. By investigation of quality of the RO penetrate based on conductivity, it can be concluded that the HF-MBR/RO followed by a RO unit *x* could penetrate provide the requirements of water for several recycle application including potable and non-potable.

4. Conclusions

In this research, the HF-MBR coupled with a RO unit has been developed and its performance for refinement of the actual wastewater of the Arak refinery Co. was studied through the investigation of eliminatin efficiency of COD, BOD_5, TSS, VSS, TDS and Turbidity. The results showed that removal amount of COD, BOD_5, TSS, VSS, TDS and Turbidity were 82%, 89%, 98%, 99%, 99% and 98%, respectively. Effect of operating variables including the inlet COD and BOD_5 concentrations and HRT on the performance of the system was studied. HRT amount of 36h results to the minimum concentrations of COD and BOD in the effluent. Therefore it can be concluded that the HF-MBR/RO could be utilized in the large scale plants of wastewater refinement. Desirable ability of the HF-MBR/RO for elimination of COD_5, BOD5, TSS, VSS, TDS and turbidity can be used for covert of the wastewater from a hazardous pollutant of environment to a water supply with potential of further reuses or for discharge to the environment more safely.

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